

The time-dependent Gutzwiller theory for multi-band Hubbard models

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Abstract. We formulate a multi-band generalisation of the time-dependent Gutzwiller theory. This approach allows for the calculation of general two-particle response functions, which are crucial for an understanding of various experiments in solid-state physics. As a first application, we study the momentum- and frequency-resolved magnetic susceptibility in a two-band Hubbard model. Like in the underlying ground-state approaches we find significant differences between the results of our method and those from a time-dependent Hartree-Fock approximation.

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1. Introduction

The study of materials with medium to strong Coulomb-interaction effects has been a central subject for experimental and theoretical solid-state physics over many years. Despite enormous efforts and significant progress in some fields, however, our theoretical toolbox is still far from satisfactory for such systems. For quite some time, theoreticians in many-particle physics have focused on relatively simple model systems, such as the Heisenberg or the single-band Hubbard models. Only in the past ten years, attention shifted towards the study of more realistic models, e.g., multi-band Hubbard models. A very important impulse in that direction came from the limit of infinite spatial dimensions ($D \rightarrow \infty$). The exact solution of Hubbard models in this limit leads to the Dynamical Mean Field Theory (DMFT), in which the original lattice model is mapped onto an effective single-impurity system that has to be solved numerically [1, 2, 3, 4, 5]. Although significant progress has been made in recent years in developing numerical techniques for the solution of the DMFT equations, it is still quite challenging and can be carried out only with limited accuracy. It is particularly difficult for the DMFT to study multi-orbital Hubbard models when the full (local) Coulomb and exchange interaction is included.

An alternative method that also relies on infinite- D techniques is the Gutzwiller variational approach. It allows for the approximate study of ground-state properties and single-particle excitations with much less numerical effort than within DMFT and has been applied in a number of works in recent years [6, 7, 8, 9, 10, 11, 12, 13, 14, 15, 16, 17, 18, 19, 20, 21, 22, 23, 24]. Another approach that leads to the same energy functional for multi-band models is the slave-boson mean field theory [25, 26, 27, 28, 29, 30].

The theoretical interpretation of a number of experiments requires the study of two-particle response functions. For example, in magnetic neutron scattering the frequency- and momentum-resolved magnetic susceptibility is measured. The textbook method for the calculation of such response functions is the random-phase approximation (RPA), which can be interpreted as a time-dependent generalisation of the Hartree-Fock (HF) theory in the small amplitude limit, i.e., where the perturbation is considered to be sufficiently small. For electronic systems with medium or strong correlation effects, however, the ground-state description of a HF theory is well known to be often inaccurate. Therefore the RPA, as the time-dependent generalisation of the HF theory, is also a questionable approach for such systems.

A time-dependent Gutzwiller theory for the calculation of two-particle response functions was developed for single-band Hubbard models by Seibold et al. [31, 32]. In recent years this approach has been applied with astonishing success to quite a number of such models and response functions [33, 34, 35, 36, 37, 38, 39, 40, 41, 42, 43]. It is the main purpose of the present work to generalise the time-dependent Gutzwiller theory for the investigation of multi-band models. A brief introduction into our method has already been given in Ref. [44]. All technical details, however, will be first presented here.

Our presentation is organised as follows: In chapters 2 and 3 we summarise the main results of the Gutzwiller variational theory for multi-band Hubbard models. In chapter 4 the reader will be reminded of the derivation which introduces the RPA as a time-dependent generalisation of the HF theory. In a very similar way the time-dependent Gutzwiller theory ('Gutzwiller RPA') is introduced in chapter 5. The general Gutzwiller RPA equations are used in chapter 6 for the calculation of response functions for Hubbard-type lattice models. As a first application we study the magnetic susceptibility in a two-band model in chapter 7. A summary and conclusions close our presentation in chapter 8. The more technical parts of our derivation are referred to four appendices.

2. Multi-band Hubbard models and Gutzwiller wave functions

We study the general class of multi-band Hubbard models

$$\hat{H} = \sum_{i \neq j; \sigma, \sigma'} t_{i,j}^{\sigma, \sigma'} \hat{c}_{i,\sigma}^\dagger \hat{c}_{j,\sigma'} + \sum_i \hat{H}_{\text{loc},i} = \hat{H}_0 + \hat{H}_{\text{loc}}. \quad (1)$$

Here, the first term describes the hopping of electrons between N spin-orbital states σ, σ' on L_s lattice sites i, j , respectively. The Hamiltonian

$$\hat{H}_{\text{loc},i} = \frac{1}{2} \sum_{\sigma_1, \sigma_2, \sigma_3, \sigma_4} U_i^{\sigma_1, \sigma_2, \sigma_3, \sigma_4} \hat{c}_{i,\sigma_1}^\dagger \hat{c}_{i,\sigma_2}^\dagger \hat{c}_{i,\sigma_3} \hat{c}_{i,\sigma_4} + \sum_{\sigma_1, \sigma_2} \epsilon_i^{\sigma_1, \sigma_2} \hat{c}_{i,\sigma_1}^\dagger \hat{c}_{i,\sigma_2} \quad (2)$$

contains all local terms, i.e., the two-particle Coulomb interactions ($\sim U_i$) and the orbital onsite-energies ($\sim \epsilon_i$). We further introduce the eigenstates $|\Gamma\rangle_i$ of $\hat{H}_{\text{loc},i}$ and the corresponding energies $E_{\Gamma,i}^{\text{loc}}$, i.e.,

$$\hat{H}_{\text{loc},i} |\Gamma\rangle_i = E_{\Gamma,i}^{\text{loc}} |\Gamma\rangle_i. \quad (3)$$

Within the Gutzwiller theory, the Hamiltonian (1) is investigated by means of the variational wave function

$$|\Psi_G\rangle = \hat{P}_G |\Psi_0\rangle = \prod_i \hat{P}_i |\Psi_0\rangle, \quad (4)$$

where $|\Psi_0\rangle$ is a normalised single-particle product state and the local Gutzwiller correlator is defined as

$$\hat{P}_i = \sum_{\Gamma, \Gamma'} \lambda_{i,\Gamma, \Gamma'} |\Gamma\rangle_{ii} \langle \Gamma'|. \quad (5)$$

For example, in case of the single-band Hubbard model

$$\hat{H}_{\text{sb}} = \sum_{i \neq j} \sum_{\sigma = \uparrow, \downarrow} t_{i,j} \hat{c}_{i,\sigma}^\dagger \hat{c}_{j,\sigma} + \sum_i U |d\rangle_{ii} \langle d|, \quad (6)$$

the local correlation operator reads

$$\begin{aligned} \hat{P}_i = & \lambda_{i,d} |d\rangle_{ii} \langle d| + \lambda_{i,\uparrow} |\uparrow\rangle_{ii} \langle \uparrow| + \lambda_{i,\downarrow} |\downarrow\rangle_{ii} \langle \downarrow| + \lambda_{i,\uparrow\downarrow} |\uparrow\downarrow\rangle_{ii} \langle \uparrow\downarrow| \\ & + \lambda_{i,\uparrow\downarrow} |\uparrow\downarrow\rangle_{ii} \langle \downarrow| + \lambda_{i,\emptyset} |\emptyset\rangle_{ii} \langle \emptyset|. \end{aligned} \quad (7)$$

Here, we introduced the atomic states $|\Gamma\rangle_i$ for doubly occupied sites $|d\rangle_i$, singly occupied sites $|\uparrow\rangle_i$ and $|\downarrow\rangle_i$, and for empty sites $|\emptyset\rangle_i$, as well as the abbreviation $\lambda_{i,\Gamma}$ for the diagonal

variational parameters $\lambda_{i,\Gamma,\Gamma}$. In terms of the fermionic operators $\hat{c}_{i,\sigma}^{(\dagger)}$, the operator (7) has the form

$$\begin{aligned} \hat{P}_i = & \lambda_{i,d}\hat{n}_{i,\uparrow}\hat{n}_{i,\downarrow} + \lambda_{i,\uparrow}\hat{n}_{i,\uparrow}(1 - \hat{n}_{i,\downarrow}) + \lambda_{i,\downarrow}\hat{n}_{i,\downarrow}(1 - \hat{n}_{i,\uparrow}) \\ & + \lambda_{i,\downarrow,\uparrow}\hat{c}_{i,\downarrow}^\dagger\hat{c}_{i,\uparrow} + \lambda_{i,\uparrow,\downarrow}\hat{c}_{i,\uparrow}^\dagger\hat{c}_{i,\downarrow} + \lambda_{i,\emptyset}(1 - \hat{n}_{i,\uparrow})(1 - \hat{n}_{i,\downarrow}) \end{aligned} \quad (8)$$

where $\hat{n}_{i,\sigma} \equiv \hat{c}_{i,\sigma}^\dagger\hat{c}_{i,\sigma}$. The correlation operator (7) is the most general Ansatz for single-band models without superconductivity. The latter would require additional terms of the form $\sim |d\rangle_{ii}\langle\emptyset|$ and $\sim |\emptyset\rangle_{ii}\langle d|$; c.f., Ref. [45].

3. Variational energy

As shown in Refs. [7, 46], the expectation value of the Hamiltonian (1) with respect to the variational wave-function (4) can be evaluated in the limit of infinite spatial dimensions. We consider the expectation values of the local Hamiltonian \hat{H}_{loc} , and the one-particle Hamiltonian \hat{H}_1 separately in sections 3.1 and 3.2. The additional constraints, which arise through the derivation in infinite dimensions are discussed in section 3.3. In section 3.4, we recall how the standard Gutzwiller energy functional for a single-band model is recovered from our general multi-band results.

3.1. Local energy

The expectation value of the local Hamiltonian (3) in infinite dimensions reads [48]

$$\langle \hat{H}_{\text{loc}} \rangle_{\Psi_G} = \sum_{\Gamma} E_{\Gamma}^{\text{loc}} m_{\Gamma,\Gamma} \equiv E_{\text{loc}} , \quad (9)$$

where

$$m_{\Gamma,\Gamma'} \equiv \langle (|\Gamma\rangle\langle\Gamma'|) \rangle_{\Psi_G} = \left\langle \left(\hat{P}^\dagger |\Gamma\rangle\langle\Gamma'| \hat{P} \right) \right\rangle_{\Psi_0} = \sum_{\tilde{\Gamma},\tilde{\Gamma}'} \lambda_{\Gamma,\tilde{\Gamma}}^* \lambda_{\Gamma',\tilde{\Gamma}'} m_{\tilde{\Gamma},\tilde{\Gamma}'}^0 \quad (10)$$

and

$$m_{\Gamma,\Gamma'}^0 \equiv \langle (|\Gamma\rangle\langle\Gamma'|) \rangle_{\Psi_0} . \quad (11)$$

To further evaluate the expectation value (11), we introduce the basis of Fock states (i.e., ‘Slater determinants’)

$$|I\rangle = \prod_{\sigma \in I} \hat{c}_{\sigma}^\dagger |0\rangle \quad (12)$$

in which certain spin-orbit states $\sigma \in I$ are occupied. Mathematically, the indices $I = (\sigma_1, \sigma_2, \dots, \sigma_n)$ are considered as ordered sets of spin-orbit states σ . Therefore we can use all standard set operations, such as $I \cup \sigma$ or $I \setminus \sigma$. In addition, we define the number of orbitals in I as $|I|$. The states $|I\rangle$ provide a basis of the local (atomic) Hilbert space. Hence, we can use them for an expansion of the eigenstates $|\Gamma\rangle$,

$$|\Gamma\rangle = \sum_I T_{I,\Gamma} |I\rangle \quad (13)$$

and write the expectation value (11) as

$$m_{\Gamma, \Gamma'}^0 = \sum_{I, I'} T_{I, \Gamma} T_{I', \Gamma'}^* m_{I, I'}^0 . \quad (14)$$

Finally, the uncorrelated expectation values of the transfer operators $|I\rangle\langle I'|$,

$$m_{I, I'}^0 \equiv \langle (|I\rangle\langle I'|) \rangle_{\Psi_0} , \quad (15)$$

can be written as the determinant

$$m_{I, I'}^0 = \begin{vmatrix} \Omega_{I, I'}^{I, I'} & -\Omega_{I, I'}^{I, J} \\ \Omega_{I, I'}^{J, I'} & \bar{\Omega}_{I, I'}^{J, J} \end{vmatrix} . \quad (16)$$

Here, $\Omega_{I, I'}$ are the matrices

$$\Omega_{I, I'} = \begin{pmatrix} C_{\sigma_1, \sigma'_1}^0 & C_{\sigma_1, \sigma'_2}^0 & \cdots & C_{\sigma_1, \sigma'_{|I'|}}^0 \\ C_{\sigma_2, \sigma'_1}^0 & C_{\sigma_2, \sigma'_2}^0 & \cdots & C_{\sigma_2, \sigma'_{|I'|}}^0 \\ \cdots & \cdots & \cdots & \cdots \\ C_{\sigma_{|I|}, \sigma'_1}^0 & C_{\sigma_{|I|}, \sigma'_2}^0 & \cdots & C_{\sigma_{|I|}, \sigma'_{|I'|}}^0 \end{pmatrix} , \quad (17)$$

in which the entries are the elements of the uncorrelated local density matrix

$$C_{\sigma, \sigma'}^0 = \langle \hat{c}_{i, \sigma}^\dagger \hat{c}_{i, \sigma'} \rangle_{\Psi_0} \quad (18)$$

that belong to the configurations $I = (\sigma_1, \dots, \sigma_{|I|})$ and $I' = (\sigma'_1, \dots, \sigma'_{|I'|})$. The matrix $\bar{\Omega}_{I, I'}^{J, J}$ in (16) is defined as

$$\bar{\Omega}_{I, I'}^{J, J} = \begin{pmatrix} 1 - C_{\sigma_1, \sigma_1}^0 & -C_{\sigma_1, \sigma_2}^0 & \cdots & -C_{\sigma_1, \sigma_{|J|}}^0 \\ -C_{\sigma_2, \sigma_1}^0 & 1 - C_{\sigma_2, \sigma_2}^0 & \cdots & -C_{\sigma_2, \sigma_{|J|}}^0 \\ \cdots & \cdots & \cdots & \cdots \\ -C_{\sigma_{|J|}, \sigma_1}^0 & -C_{\sigma_{|J|}, \sigma_2}^0 & \cdots & 1 - C_{\sigma_{|J|}, \sigma_{|J|}}^0 \end{pmatrix} , \quad (19)$$

with $\sigma_i \in J \equiv (1, \dots, N) \setminus (I \cup I')$.

In applications of the Gutzwiller theory to multi-band systems it would be quite cumbersome to evaluate the determinants (16) if the local density matrix (18) is non-diagonal. Fortunately, we are free to choose the local orbital basis in a way that suits us best. Therefore, we introduce an orbital basis, defined by (local) operators $\hat{h}_\gamma^{(\dagger)}$, for which

$$C_{\gamma, \gamma'}^0 = \bar{C}_{\gamma, \gamma'}^0 \equiv \delta_{\gamma, \gamma'} \langle \hat{h}_{i, \gamma}^\dagger \hat{h}_{i, \gamma'} \rangle_{\Psi_0} \equiv n_\gamma^0 . \quad (20)$$

With such a basis the expectation value (16) has the simple form

$$m_{I, I'}^0 = \delta_{I, I'} \prod_{\gamma \in I} n_\gamma^0 \prod_{\gamma \notin I} (1 - n_\gamma^0) . \quad (21)$$

Note that, for simplicity, we always use the same variable I for configuration states of the form (12) irrespective of the underlying orbital basis (e.g., $\hat{c}_\sigma^{(\dagger)}$ in (12) or $\hat{h}_\gamma^{(\dagger)}$ in (21)).

As will be shown in chapter 5, the time-dependent Gutzwiller theory requires to calculate the first and second derivatives of the energy with respect to all elements of the local density matrix, including the non-diagonal terms. Since the local density matrix

enters the energy functional solely through matrices of the form (16) we only need to expand these matrices with respect to small perturbations

$$C_{\gamma,\gamma'}^0 = \bar{C}_{\gamma,\gamma'}^0 + \delta C_{\gamma,\gamma'}^0 \quad (22)$$

up to second order in $\delta C_{\gamma,\gamma'}^0$ around the diagonal ground-state matrix (20). This expansion is explicitly carried out in Appendix A.

In our derivation of the ground-state energy all local onsite energies were considered as part of the local Hamiltonian (2). For later use, however, we also need an expression for the expectation value of a general local one-particle Hamiltonian

$$\hat{H}_{\text{onsite}} = \sum_{\sigma,\sigma'} \epsilon^{\sigma,\sigma'} \hat{c}_{\sigma}^{\dagger} \hat{c}_{\sigma'} \quad (23)$$

This expectation value is given as

$$\langle \hat{H}_{\text{onsite}} \rangle_{\Psi_G} = \sum_{\sigma,\sigma'} \epsilon^{\sigma,\sigma'} C_{\sigma,\sigma'}^c \quad (24)$$

where

$$C_{\sigma,\sigma'}^c = \sum_{\Gamma_1, \Gamma_2, \Gamma_3, \Gamma_4} \lambda_{\Gamma_2, \Gamma_1}^* \lambda_{\Gamma_3, \Gamma_4} \langle \Gamma_2 | \hat{c}_{\sigma}^{\dagger} \hat{c}_{\sigma'} | \Gamma_3 \rangle m_{\Gamma_1, \Gamma_4}^0 \quad (25)$$

is the ‘correlated’ local density matrix.

3.2. Kinetic energy

The expectation value of a hopping term in \hat{H}_0 , Eq. (1), is given as

$$\langle \hat{c}_{i,\sigma_1}^{\dagger} \hat{c}_{j,\sigma_2} \rangle_{\Psi_G} = \sum_{\sigma'_1, \sigma'_2} q_{i,\sigma_1}^{\sigma'_1} \left(q_{j,\sigma_2}^{\sigma'_2} \right)^* \langle \hat{c}_{i,\sigma'_1}^{\dagger} \hat{c}_{j,\sigma'_2} \rangle_{\Psi_0} \quad (26)$$

where we have introduced the (local) renormalisation matrix [48]

$$q_{\sigma}^{\sigma'} = \sum_{\Gamma_1, \dots, \Gamma_4} \lambda_{\Gamma_2, \Gamma_1}^* \lambda_{\Gamma_3, \Gamma_4} \langle \Gamma_2 | \hat{c}_{\sigma}^{\dagger} | \Gamma_3 \rangle \sum_{I_1, I_4} T_{I_1, \Gamma_1} T_{I_4, \Gamma_4}^* H_{I_1, I_4}^{\sigma'} \quad (27)$$

The matrix $H_{I_1, I_4}^{\sigma'}$ contains three different contributions depending on whether the index σ' is an element of $I_1 \cap I_4$, $I_4 \setminus (I_1 \cap I_4)$, or $J = (1, \dots, N) \setminus (I_1 \cup I_4)$. With the abbreviation $f_{\sigma, I} \equiv \langle I | \hat{c}_{\sigma}^{\dagger} \hat{c}_{\sigma} | I \rangle$ we can write $H_{I_1, I_4}^{\sigma'}$ as

$$\begin{aligned} H_{I_1, I_4}^{\sigma'} &\equiv (1 - f_{\sigma', I_1}) \langle I_4 | \hat{c}_{\sigma'} | I_4 \cup \sigma' \rangle m_{I_1, I_4 \cup \sigma'}^0 \\ &+ \langle I_1 \setminus \sigma' | \hat{c}_{\sigma'} | I_1 \rangle \left(f_{\sigma', I_4} m_{I_1 \setminus \sigma', I_4}^0 + (1 - f_{\sigma', I_4}) m_{I_1 \setminus \sigma', I_4}^{0; \sigma'} \right) \end{aligned} \quad (28)$$

The expectation value $m_{I_1 \setminus \sigma', I_4}^{0; \sigma'}$ in (28) has the same form as the one in (16), except that the index J has to be replaced by $J \setminus \sigma'$. In case of a diagonal local density matrix one finds

$$H_{I_1, I_4}^{\sigma'} = \delta_{I_1 \setminus \sigma', I_4} \langle I_1 \setminus \sigma' | \hat{c}_{\sigma'} | I_1 \rangle \frac{m_{I_4, I_4}^0}{1 - C_{\sigma', \sigma'}^0} \quad (29)$$

in agreement with results derived earlier [7]. Note that, in general, the renormalisation matrix is *not* Hermitian, i.e., it is

$$q_{\sigma}^{\sigma'} \neq (q_{\sigma'}^{\sigma})^* . \quad (30)$$

Using (26), the expectation value of the one particle Hamiltonian \hat{H}_0 can be written as

$$\langle \hat{H}_0 \rangle_{\Psi_G} = L_s \sum_{\sigma_1, \sigma_2, \sigma'_1, \sigma'_2} q_{\sigma_1}^{\sigma'_1} (q_{\sigma_2}^{\sigma'_2})^* E_{\sigma_1, \sigma_2, \sigma'_1, \sigma'_2} \quad (31)$$

where we introduced the tensor

$$E_{\sigma_1, \sigma_2, \sigma'_1, \sigma'_2} \equiv \frac{1}{L_s} \sum_{i \neq j} t_{i,j}^{\sigma_1, \sigma_2} \langle \hat{c}_{i, \sigma'_1}^\dagger \hat{c}_{j, \sigma'_2} \rangle_{\Psi_0} . \quad (32)$$

3.3. Physical constraints

As it turns out through the evaluation of expectation values in infinite dimensions, the variational parameters $\lambda_{\Gamma, \Gamma'}$ need to obey certain local constraints. These are

$$\langle \hat{P}^\dagger \hat{P} \rangle_{\Psi_0} = 1 , \quad (33)$$

$$\langle \hat{c}_\sigma^\dagger \hat{P}^\dagger \hat{P} \hat{c}_{\sigma'} \rangle_{\Psi_0} = \langle \hat{c}_\sigma^\dagger \hat{c}_{\sigma'} \rangle_{\Psi_0} = C_{\sigma, \sigma'}^0 . \quad (34)$$

Note that moving the operator $\hat{P}^\dagger \hat{P}$ relative to \hat{c}_σ^\dagger or $\hat{c}_{\sigma'}$ in (34) would not alter the whole set of constraints. With the explicit form (5) of the correlation operator \hat{P} , the constraints read

$$1 = \sum_{\Gamma, \Gamma_1, \Gamma_2} \lambda_{\Gamma, \Gamma_1}^* \lambda_{\Gamma, \Gamma_2} m_{\Gamma_1, \Gamma_2}^0 , \quad (35)$$

$$C_{\sigma, \sigma'}^0 = \sum_{\Gamma, \Gamma', \Gamma_1, \Gamma_2, \Gamma_3} \lambda_{\Gamma_2, \Gamma_1}^* \lambda_{\Gamma_2, \Gamma_3} \langle \Gamma | \hat{c}_\sigma^\dagger | \Gamma_1 \rangle \times \langle \Gamma_3 | \hat{c}_{\sigma'} | \Gamma' \rangle m_{\Gamma, \Gamma'}^0 . \quad (36)$$

3.4. Recovery of the ‘standard’ single-band energy functional

In case of a single-band model, the atomic eigenstates $|\Gamma\rangle$ coincide with the configuration states $|I\rangle$. If we assume a translationally invariant ground state and the most general form of a local density matrix [48]

$$C^0 = \begin{pmatrix} \langle \hat{c}_\uparrow^\dagger \hat{c}_\uparrow \rangle_{\Psi_0} & \langle \hat{c}_\uparrow^\dagger \hat{c}_\downarrow \rangle_{\Psi_0} \\ \langle \hat{c}_\downarrow^\dagger \hat{c}_\uparrow \rangle_{\Psi_0} & \langle \hat{c}_\downarrow^\dagger \hat{c}_\downarrow \rangle_{\Psi_0} \end{pmatrix} = \begin{pmatrix} n_\uparrow^0 & \Delta_{\uparrow, \downarrow}^0 \\ \Delta_{\downarrow, \uparrow}^0 & n_\downarrow^0 \end{pmatrix} , \quad (37)$$

where $(\Delta_{\downarrow, \uparrow}^0)^* = \Delta_{\uparrow, \downarrow}^0 \equiv \Delta^0$, we find

$$m_{\emptyset, \emptyset}^0 = (1 - n_\uparrow^0)(1 - n_\downarrow^0) - |\Delta^0|^2 , \quad (38)$$

$$m_{\sigma, \sigma}^0 = n_\sigma^0(1 - n_{\bar{\sigma}}^0) + |\Delta^0|^2 , \quad (39)$$

$$m_{\sigma, \bar{\sigma}}^0 = \Delta_{\sigma, \bar{\sigma}}^0 , \quad (40)$$

$$m_{d, d}^0 = n_\uparrow^0 n_\downarrow^0 - |\Delta^0|^2 , \quad (41)$$

for those of the expectation values (15) which are finite. Here we used the notation

$$\bar{\uparrow} = \downarrow \quad \text{and} \quad \bar{\downarrow} = \uparrow . \quad (42)$$

As a consequence, the expectation value of the local Coulomb interaction in (6) reads

$$\sum_i U \langle |d\rangle_{ii} \langle d| \rangle_{\Psi_G} = L_s U |\lambda_d|^2 m_{d,d}^0. \quad (43)$$

For the single-band model with the correlation operator (7) and the local density matrix (37) the elements of the renormalisation matrix have the form

$$q_\sigma^\sigma = \lambda_\sigma^* \lambda_\emptyset (1 - n_\sigma^0) + \lambda_d^* \lambda_{\bar{\sigma}} n_{\bar{\sigma}}^0 + (\lambda_d^* \lambda_{\bar{\sigma},\sigma} + \lambda_{\sigma,\bar{\sigma}}^* \lambda_\emptyset) \Delta_{\bar{\sigma},\sigma}^0, \quad (44)$$

$$q_{\bar{\sigma}}^{\bar{\sigma}} = \Delta_{\sigma,\bar{\sigma}}^0 (\lambda_\sigma^* \lambda_\emptyset - \lambda_d^* \lambda_{\bar{\sigma}}) - \lambda_d^* \lambda_{\bar{\sigma},\sigma} n_\sigma^0 + \lambda_{\sigma,\bar{\sigma}}^* \lambda_\emptyset (1 - n_\sigma^0). \quad (45)$$

Finally, the constraints in this case are given as

$$\begin{aligned} 1 &= |\lambda_\emptyset|^2 m_{\emptyset,\emptyset}^0 + |\lambda_d|^2 m_{d,d}^0 + (|\lambda_\uparrow|^2 + |\lambda_{\downarrow,\uparrow}|^2) m_{\uparrow,\uparrow}^0 + (|\lambda_\downarrow|^2 + |\lambda_{\uparrow,\downarrow}|^2) m_{\downarrow,\downarrow}^0 \\ &\quad + (\lambda_{\downarrow,\uparrow}^* \lambda_\downarrow + \lambda_\uparrow^* \lambda_{\uparrow,\downarrow}) \Delta_{\uparrow,\downarrow}^0 + (\lambda_{\uparrow,\downarrow}^* \lambda_\uparrow + \lambda_\downarrow^* \lambda_{\downarrow,\uparrow}) \Delta_{\downarrow,\uparrow}^0, \end{aligned} \quad (46)$$

$$n_\sigma^0 = (|\lambda_\sigma|^2 + |\lambda_{\sigma,\bar{\sigma}}|^2) m_{d,d}^0 + |\lambda_\emptyset|^2 m_{\sigma,\sigma}^0, \quad (47)$$

$$\Delta_{\sigma,\bar{\sigma}}^0 = -(\lambda_{\sigma,\bar{\sigma}}^* \lambda_\sigma + \lambda_{\bar{\sigma},\sigma}^* \lambda_{\bar{\sigma}}) m_{d,d}^0 + |\lambda_\emptyset|^2 \Delta_{\sigma,\bar{\sigma}}^0. \quad (48)$$

As mentioned before, it is possible to overcome the complications that arise from a non-diagonal local density matrix by a simple transformation

$$\hat{h}_\gamma^\dagger = \sum_\sigma u_{\sigma,\gamma} \hat{c}_\sigma^\dagger \quad (49)$$

to a new orbital basis for which the local density matrix is diagonal by definition,

$$\begin{pmatrix} \langle \hat{h}_1^\dagger \hat{h}_1 \rangle_{\Psi_0} & \langle \hat{h}_1^\dagger \hat{h}_2 \rangle_{\Psi_0} \\ \langle \hat{h}_2^\dagger \hat{h}_1 \rangle_{\Psi_0} & \langle \hat{h}_2^\dagger \hat{h}_2 \rangle_{\Psi_0} \end{pmatrix} = \begin{pmatrix} \tilde{n}_1^0 & 0 \\ 0 & \tilde{n}_2^0 \end{pmatrix}. \quad (50)$$

In this new basis the constraints have the rather simple form

$$1 = \tilde{\lambda}_\emptyset^2 m_{\emptyset,\emptyset}^0 + \tilde{\lambda}_1^2 m_{1,1}^0 + \tilde{\lambda}_2^2 m_{2,2}^0 + \tilde{\lambda}_d^2 m_{d,d}^0, \quad (51)$$

$$\tilde{n}_\gamma^0 = \tilde{\lambda}_\gamma^2 m_{\gamma,\gamma}^0 + \tilde{\lambda}_d^2 m_{d,d}^0. \quad (52)$$

where the non-diagonal constraints are automatically fulfilled by working with a diagonal correlation operator ($\tilde{\lambda}_{1,2} = 0$). Equations (51)-(52) can be readily solved by introducing the expectation values

$$\tilde{m}_d \equiv \tilde{\lambda}_d^2 m_{d,d}^0, \quad (53)$$

$$\tilde{m}_\emptyset \equiv \tilde{\lambda}_\emptyset^2 m_{\emptyset,\emptyset}^0 = 1 - \tilde{n}_1^0 - \tilde{n}_2^0 + \tilde{m}_d, \quad (54)$$

$$\tilde{m}_\gamma \equiv \tilde{\lambda}_\gamma^2 m_{\gamma,\gamma}^0 = \tilde{n}_\gamma^0 - \tilde{m}_d, \quad (55)$$

which leaves us with only one variational parameter, the expectation value \tilde{m}_d for a double occupancy. The resulting renormalisation matrix is then diagonal and its elements have the well known form [49, 50, 51]

$$q_\gamma \equiv q_\gamma^\gamma = \tilde{\lambda}_\emptyset \tilde{\lambda}_\gamma (1 - \tilde{n}_\gamma^0) + \tilde{\lambda}_d \tilde{\lambda}_{\bar{\gamma}} \tilde{n}_{\bar{\gamma}}^0 \frac{1}{\sqrt{\tilde{n}_\gamma^0 (1 - \tilde{n}_\gamma^0)}} \left(\sqrt{\tilde{m}_\emptyset \tilde{m}_\gamma} + \sqrt{\tilde{m}_d \tilde{m}_{\bar{\gamma}}} \right). \quad (56)$$

Hence, the single-particle energy (31) is given as

$$E_0 = \sum_\gamma q_\gamma^2 \sum_{i \neq j} t_{i,j} \langle \hat{h}_{i,\gamma}^\dagger \hat{h}_{j,\gamma} \rangle_{\Psi_0} \quad (57)$$

where we used the orthonormality relation

$$\sum_{\sigma} u_{\sigma,\gamma} u_{\sigma,\gamma'}^* = \delta_{\gamma,\gamma'} . \quad (58)$$

In order to formally show the equivalence of both approaches we write the parameters $\lambda_{\Gamma,\Gamma'}$ in (7) as

$$\lambda_{\emptyset} = \tilde{\lambda}_{\emptyset} , \quad (59)$$

$$\lambda_d = \tilde{\lambda}_d , \quad (60)$$

$$\lambda_{\sigma,\sigma'} = \sum_{\gamma} u_{\sigma,\gamma} u_{\sigma',\gamma}^* \tilde{\lambda}_{\gamma} . \quad (61)$$

With these relations it is easy to show that the constraints (46)-(48) are indeed fulfilled. For example, the first constraint (46) can be written as

$$1 = \lambda_{\emptyset}^2 m_{\emptyset,\emptyset}^0 + \lambda_d^2 m_{d,d}^0 + \sum_{\sigma,\sigma',\sigma''} \lambda_{\sigma'',\sigma}^* \lambda_{\sigma'',\sigma'} m_{\sigma,\sigma'}^0 . \quad (62)$$

If we use

$$m_{\sigma,\sigma'}^0 = \sum_{\gamma} u_{\sigma,\gamma}^* u_{\sigma',\gamma} m_{\gamma}^0 \quad (63)$$

and the orthonormality relation (58) we readily find that (62) is indeed solved by the parameters (59)-(61). In the same way one can show the equivalence of the ground-state energy functionals.

4. The Time-Dependent Hartree-Fock Approximation

The approximation most frequently applied to two-particle Green's functions is the 'random-phase approximation' (RPA). This approach can be derived in various ways, e.g., by an equation of motion technique or in diagrammatic perturbation theory [52]. In this section, we use a different derivation which introduces the RPA as a time-dependent generalisation of the Hartree-Fock theory; see, e.g., Refs. [53, 54]. If derived in this way, the approach can be generalised quite naturally in order to formulate a time-dependent Gutzwiller theory. This will be the subject of chapter 5.

4.1. The Hartree-Fock approximation

In the Hartree-Fock approximation a single-particle product wave function $|\Psi_0\rangle$ is used in order to investigate the ground-state properties of a many-particle system. Note that such wave functions are included in the Gutzwiller variational space by setting $\lambda_{i,\Gamma,\Gamma'} = \delta_{\Gamma,\Gamma'}$. The expectation value of a many-particle Hamiltonian with respect to a Hartree-Fock wave function is a function of the single-particle density matrix. For example, for the Hamiltonian (1) it reads

$$\begin{aligned} E^{\text{HF}}(\tilde{\rho}) &\equiv \langle \hat{H} \rangle_{\Psi_0} \\ &= \sum_{i \neq j; \sigma, \sigma'} t_{i,j}^{\sigma, \sigma'} \rho_{(j\sigma'), (i\sigma)} + \sum_{i; \sigma_1, \sigma_2} \epsilon_i^{\sigma_1, \sigma_2} \rho_{(i\sigma_2), (i\sigma_1)} + \sum_i E_{\text{loc},i}^{\text{HF}}(\tilde{\rho}) \end{aligned} \quad (64)$$

where

$$\rho_{(j\sigma'),(i\sigma)} \equiv \langle \hat{c}_{i,\sigma}^\dagger \hat{c}_{j,\sigma'} \rangle_{\Psi_0} \quad (65)$$

are the elements of the single-particle density matrix $\tilde{\rho}$ and

$$E_{\text{loc},i}^{\text{HF}}(\tilde{\rho}) = \frac{1}{2} \sum_{\substack{\sigma_1, \sigma_2, \\ \sigma_3, \sigma_4}} U_i^{\sigma_1, \sigma_2, \sigma_3, \sigma_4} \left[\rho_{(i\sigma_4), (i\sigma_1)} \rho_{(i\sigma_3), (i\sigma_2)} - \rho_{(i\sigma_3), (i\sigma_1)} \rho_{(i\sigma_4), (i\sigma_2)} \right] \quad (66)$$

is the expectation value of the Coulomb interaction in the Hamiltonian (2). Note that it will be more convenient both in the time-dependent Hartree-Fock and Gutzwiller theory, to use a different order of subscripts in the definition (65) of density matrices than, e.g., in section 3 or in previous work on the Gutzwiller theory.

To keep notations simple, we use the abbreviations $v \equiv (i, \sigma)$ for local single-particle states and $Y = (v, v')$ for pairs of these indices. For example, the elements of $\tilde{\rho}$ can then be written as

$$\rho_Y = \rho_{v_1, v_2} = \rho_{(i_1, \sigma_1), (i_2, \sigma_2)} \quad (67)$$

With these new notations, the Hartree-Fock energy (64) reads

$$\begin{aligned} E^{\text{HF}}(\tilde{\rho}) &= \sum_{v_1, v_2} \varepsilon_{v_1, v_2} \rho_{v_2, v_1} + \frac{1}{2} \sum_{\substack{v_1, v_2 \\ v_3, v_4}} \rho_{v_4, v_1} W_{(v_1, v_4), (v_3, v_2)} \rho_{v_3, v_2} \\ &= \sum_Y \varepsilon_Y \rho_{\bar{Y}} + \frac{1}{2} \sum_{Y, Y'} \rho_{\bar{Y}} W_{Y, Y'} \rho_{Y'} \end{aligned} \quad (68)$$

where

$$\varepsilon_{(i\sigma_1), (j\sigma_2)} \equiv t_{i,j}^{\sigma_1, \sigma_2} + \delta_{i,j} \epsilon_i^{\sigma_1, \sigma_2} \quad (69)$$

and

$$W_{(v_1, v_4), (v_3, v_2)} \equiv U_i^{\sigma_1, \sigma_2, \sigma_3, \sigma_4} - U_i^{\sigma_1, \sigma_2, \sigma_4, \sigma_3} \quad (70)$$

for indices $v_k = (i, \sigma_k)$ that belong to the same lattice site i . Further, we introduced the ‘inverse’ index $\bar{Y} \equiv (v', v)$ for $Y = (v, v')$. Note the symmetries

$$W_{(v_1, v_4), (v_3, v_2)} = W_{(v_2, v_3), (v_4, v_1)} = -W_{(v_1, v_3), (v_4, v_2)} \quad (71)$$

which will be employed in the following section.

The energy functional (68) has to be minimised with respect to all density matrices which belong to a single-particle product state. Such matrices are idempotent, i.e., they obey the matrix equation

$$\tilde{\rho}^2 = \tilde{\rho} \quad (72)$$

If one imposes this constraint via a Lagrange parameter matrix $\tilde{\eta}$ with elements $\eta_{v, v'}$, the following equation has to be solved

$$\frac{\partial}{\partial \rho_{v, v'}} \left[E^{\text{HF}}(\tilde{\rho}) - \text{tr} \left(\tilde{\eta} (\tilde{\rho}^2 - \tilde{\rho}) \right) \right] = 0 \quad (73)$$

This condition leads to

$$\tilde{h}(\tilde{\rho}) + \tilde{\eta} - \tilde{\eta} \tilde{\rho} - \tilde{\rho} \tilde{\eta} = 0 \quad (74)$$

where we introduced the matrix $\tilde{h}(\tilde{\rho})$ with the elements

$$h_Y(\tilde{\rho}) = \frac{\partial}{\partial \rho_Y} E^{\text{HF}}(\tilde{\rho}) = \varepsilon_Y + \sum_{Y'} W_{Y,Y'} \rho_{Y'} . \quad (75)$$

Equation (74) is solved if $\tilde{\rho}$ satisfies both (72) and

$$[\tilde{h}(\tilde{\rho}), \tilde{\rho}] = 0 . \quad (76)$$

Starting with a certain density matrix $\tilde{\rho}$ we can introduce the ‘Hartree-Fock’ basis

$$|\alpha\rangle = \sum_v u_{v,\alpha} |v\rangle \quad (77)$$

of states which diagonalise the Hamilton matrix $\tilde{h}(\tilde{\rho})$, i.e.,

$$\sum_{v'} h_{v,v'}(\tilde{\rho}) u_{v',\alpha} = E_\alpha u_{v,\alpha} . \quad (78)$$

Equation (76) is then solved by setting

$$\rho_{\alpha,\alpha'} = \delta_{\alpha,\alpha'} \Theta(E_F - E_\alpha) \quad (79)$$

where the Fermi energy E_F is determined by the total number of particles

$$N = \sum_\alpha \Theta(E_F - E_\alpha) . \quad (80)$$

The density matrix (79) has to be reinserted into (68),(75) until self-consistency is reached. We denote the solution of these equations as $\tilde{\rho}^0$ and introduce the corresponding Hamilton matrix

$$\tilde{h}^0 \equiv \tilde{h}(\tilde{\rho}^0) . \quad (81)$$

4.2. Equation of Motion for the Density Matrix

We consider two-particle Green’s functions of the form

$$\begin{aligned} G_{(v_2,v_1),(v_3,v_4)}(t-t') &\equiv \langle \langle \hat{c}_{v_1}^\dagger(t) \hat{c}_{v_2}(t); \hat{c}_{v_3}^\dagger(t') \hat{c}_{v_4}(t') \rangle \rangle \\ &\equiv -i\Theta(t-t') \langle \Phi_0 | [\hat{c}_{v_1}^\dagger(t) \hat{c}_{v_2}(t), \hat{c}_{v_3}^\dagger(t') \hat{c}_{v_4}(t')] | \Phi_0 \rangle , \end{aligned} \quad (82)$$

where $|\Phi_0\rangle$ is the exact ground state of our multi-band Hubbard Hamiltonian (1), and $\hat{c}_v^{(\dagger)}(t)$ is the Heisenberg representation of the operators $\hat{c}_v^{(\dagger)}$ with respect to \hat{H} . As shown in most textbooks on many-particle physics, the Green’s functions (82) naturally arise in ‘linear-response theory’ because they describe the time-dependent changes

$$\begin{aligned} \delta \langle \hat{c}_{v_1}^\dagger \hat{c}_{v_2} \rangle_t &\equiv \langle \hat{c}_{v_1}^\dagger \hat{c}_{v_2} \rangle_t - \langle \hat{c}_{v_1}^\dagger \hat{c}_{v_2} \rangle_{-\infty} \equiv \delta \rho_{v_2,v_1}(t) \\ &= \sum_{v_3,v_4} \int_{-\infty}^{\infty} dt' G_{(v_2,v_1),(v_3,v_4)}(t-t') f_{v_3,v_4}(t') \end{aligned} \quad (83)$$

of the density matrix $\tilde{\rho}$ in the presence of a small time-dependent perturbation

$$\hat{V}_f(t) = \sum_{v,v'} f_{v,v'}(t) \hat{c}_v^\dagger \hat{c}_{v'} \quad (84)$$

added to \hat{H} [55, 56, 57]. After a Fourier transformation and using again the abbreviation $Y = (v, v')$, Eq. (83) reads

$$\delta\rho_Y(\omega) = \sum_{Y'} G_{Y,Y'}(\omega) f_{Y'}(\omega) \quad (85)$$

with

$$G_{Y,Y'}(\omega) \equiv \int_{-\infty}^{\infty} d\tau G_{Y,Y'}(\tau) e^{i\omega\tau}, \quad (86)$$

and $f_Y(\omega)$ and $\delta\rho_Y(\omega)$ defined accordingly.

Ideally, we would like to calculate the time dependence of the density matrix

$$\rho_{v',v}(t) \equiv \langle \Psi(t) | \hat{c}_v^\dagger \hat{c}_{v'} | \Psi(t) \rangle, \quad (87)$$

where $|\Psi(t)\rangle$ is the exact solution of the time-dependent Schrödinger equation for the Hamiltonian

$$\hat{H}(t) = \hat{H} + \hat{V}_f(t). \quad (88)$$

The expectation value (87) obeys the Heisenberg equation

$$-i\dot{\rho}_{v',v}(t) = \langle \Psi(t) | [\hat{H}, \hat{c}_v^\dagger \hat{c}_{v'}] | \Psi(t) \rangle, \quad (89)$$

which contains the commutator

$$\begin{aligned} [\hat{H}(t), \hat{c}_v^\dagger \hat{c}_{v'}] &= \sum_{v_1} (\varepsilon_{v_1,v} + f_{v_1,v}(t)) \hat{c}_{v_1}^\dagger \hat{c}_{v'} - \sum_{v_1} (\varepsilon_{v',v_1} + f_{v',v_1}(t)) \hat{c}_v^\dagger \hat{c}_{v_1} \\ &+ \frac{1}{2} \sum_{v_1, v_2, v_3} \left(W_{(v_1, v_3), (v, v_2)} \hat{c}_{v_1}^\dagger \hat{c}_{v_2}^\dagger \hat{c}_{v'} \hat{c}_{v_3} + W_{(v_1, v_2), (v_3, v')} \hat{c}_{v_1}^\dagger \hat{c}_v^\dagger \hat{c}_{v_2} \hat{c}_{v_3} \right). \end{aligned} \quad (90)$$

In the time-dependent Hartree-Fock approximation, it is assumed that the solution $|\Psi(t)\rangle$ of the Schrödinger equation at any time t is approximately given by a single-particle product wave function. In this case, the expectation value of the commutator (90) can be evaluated by means of Wick's theorem. This leads to the equation of motion

$$i\dot{\tilde{\rho}}(t) = [\tilde{h}(\tilde{\rho}(t)) + \tilde{f}(t), \tilde{\rho}(t)] \quad (91)$$

for $\tilde{\rho}(t)$, where the matrix $\tilde{h}(\tilde{\rho})$ has been introduced in (75). Equations (75) and (91) will be crucial also for our formulation of a time-dependent Gutzwiller theory in chapter 5.

4.3. Expansion for Weak Perturbations

We are only interested in cases where

$$\hat{V}_f(t) \rightarrow \delta\hat{V}_f(t) = \sum_{v, v'} \delta f_{v, v'}(t) \hat{c}_v^\dagger \hat{c}_{v'} \quad (92)$$

is a weak perturbation to the time-independent Hamiltonian \hat{H} . In this case, the density matrix $\tilde{\rho}(t)$ and the Hamilton matrix $\tilde{h}(t)$ are given as

$$\tilde{\rho}(t) \approx \tilde{\rho}^0 + \delta\tilde{\rho}(t), \quad (93)$$

$$\tilde{h}(t) \approx \tilde{h}^0 + \delta\tilde{h}(t), \quad (94)$$

where $\delta\tilde{\rho}(t)$ describes a ‘small’ time-dependent perturbation around the ground-state density matrix $\tilde{\rho}^0$, and

$$h_Y^0 = \varepsilon_Y + \sum_{Y'} W_{Y,Y'} \rho_{Y'}^0, \quad (95)$$

$$\delta h_Y(t) = \sum_{Y'} W_{Y,Y'} \delta \rho_{Y'}(t). \quad (96)$$

With the expansion (93)-(94), the equation of motion (91) becomes

$$0 = [\tilde{h}^0, \tilde{\rho}^0], \quad (97)$$

$$i\delta\dot{\tilde{\rho}}(t) = [\tilde{h}^0, \delta\tilde{\rho}(t)] + [\delta\tilde{h}(t) + \delta\tilde{f}(t), \tilde{\rho}^0]. \quad (98)$$

These equations have to be solved for density matrices $\tilde{\rho}(t)$ that obey the matrix equation (72). After applying the expansion (93), Eq. (72) reads (to leading order in $\delta\tilde{\rho}(t)$)

$$\tilde{\rho}^0 = (\tilde{\rho}^0)^2, \quad (99)$$

$$\delta\tilde{\rho}(t) = \tilde{\rho}^0 \delta\tilde{\rho}(t) + \delta\tilde{\rho}(t) \tilde{\rho}^0. \quad (100)$$

Note that Eqs. (97),(99) just recover the time-independent Hartree-Fock equations derived in section 4.1.

4.4. Random -phase approximation (RPA) equations

Mathematically, the density matrix is a projector onto ‘hole’-states, $\tilde{\rho}_h \equiv \tilde{\rho}^0$. In addition, we define the projector onto ‘particle’-states as

$$\tilde{\rho}_p \equiv 1 - \tilde{\rho}^0. \quad (101)$$

With these two operators, we can decompose all matrices into their four components

$$\delta\tilde{\rho}^{vw}(t) \equiv \tilde{\rho}_v \delta\tilde{\rho}(t) \tilde{\rho}_w, \quad (102)$$

$$\delta\tilde{f}^{vw}(t) \equiv \tilde{\rho}_v \delta\tilde{f}(t) \tilde{\rho}_w, \quad (103)$$

$$\tilde{h}^{0;vw} \equiv \tilde{\rho}_v \tilde{h}^0 \tilde{\rho}_w, \quad (104)$$

where $v, w \in \{p, h\}$. Note that $\tilde{h}^{0;vw}$ has the elements

$$h_{\alpha,\alpha'}^{0;vw} = \delta_{v,w} \delta_{\alpha,\alpha'} E_\alpha. \quad (105)$$

An evaluation of the condition (100) for the components $\delta\tilde{\rho}^{vw}(t)$ yields

$$\delta\tilde{\rho}^{vw}(t) = \delta\tilde{\rho}^{vv}(t) + \delta\tilde{\rho}^{vv}(t) \delta\tilde{\rho}^{vw}(t) + \delta\tilde{\rho}^{vw}(t) \delta\tilde{\rho}^{ww}(t), \quad (106)$$

and

$$\delta\tilde{\rho}^{ww}(t) = 0 \quad (107)$$

where $v \neq w$. Hence the components $\delta\tilde{\rho}^{pp}(t)$ and $\delta\tilde{\rho}^{hh}(t)$ can be neglected in the following compared to the leading fluctuations $\delta\tilde{\rho}^{hp}(t)$ and $\delta\tilde{\rho}^{ph}(t)$.

We express the time-dependent quantities $\delta\tilde{\rho}^{vw}(t)$ and $\delta\tilde{f}^{vw}(t)$ by their respective Fourier transforms $\delta\tilde{\rho}^{vw}(\omega)$ and $\delta\tilde{f}^{vw}(\omega)$. The equation of motion (98) then leads to

$$+ \omega \delta\tilde{\rho}_{\alpha_1, \alpha_2}^{vw}(\omega) = (E_{\alpha_1} - E_{\alpha_2}) \delta\tilde{\rho}_{\alpha_1, \alpha_2}^{vw}(\omega) \pm (\delta h_{\alpha_1, \alpha_2}^{vw}(\omega) + \delta f_{\alpha_1, \alpha_2}^{vw}(\omega)) \quad (108)$$

where the plus and minus signs correspond to $vw = \text{ph}$ and $vw = \text{hp}$, respectively. With the abbreviation $A = (\alpha_1, \alpha_2)$ for pairs of indices α we find

$$\delta h_A^{vw}(\omega) = - \sum_{A'} U_{A,A'} (\delta \rho_{A'}^{vw}(\omega) + \delta \rho_{A'}^{wv}(\omega)) . \quad (109)$$

Here, the elements of the matrix \tilde{U} are given as

$$U_{A,A'} = U_{(\alpha_1, \alpha_2), (\alpha'_1, \alpha'_2)} \equiv - \sum_{\substack{v_1, v_2, \\ v'_1, v'_2}} u_{v_1, \alpha_1}^* u_{v_2, \alpha_2} W_{(v_1, v_2), (v'_1, v'_2)} u_{v'_1, \alpha'_1} u_{v'_2, \alpha'_2}^* . \quad (110)$$

The coefficients $u_{v, \alpha}$ in (110) have been introduced in Eq. (77) and determine the solutions $|\alpha\rangle$ of the Hartree-Fock equations. Equations (108) and (109) then yield

$$\left[(\omega - \tilde{E}) \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix} + \tilde{U} \right] \begin{pmatrix} \delta \tilde{\rho}^{\text{ph}}(\omega) \\ \delta \tilde{\rho}^{\text{hp}}(\omega) \end{pmatrix} = \begin{pmatrix} \delta \tilde{f}^{\text{ph}}(\omega) \\ \delta \tilde{f}^{\text{hp}}(\omega) \end{pmatrix} . \quad (111)$$

with a matrix \tilde{E} defined as

$$E_{A,A'} = E_{(\alpha_1, \alpha_2), (\alpha'_1, \alpha'_2)} = \delta_{\alpha_1, \alpha'_1} \delta_{\alpha_2, \alpha'_2} (E_{\alpha_1} - E_{\alpha_2}) . \quad (112)$$

By comparing Eqs. (111) and (85) we find

$$\tilde{G}^{-1}(\omega) = \left[(\omega + i\delta - \tilde{E}) \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix} + \tilde{U} \right] \quad (113)$$

for the inverse of the two-particle Green's function

$$\begin{aligned} G_{A,A'}(\omega) &= G_{(\alpha_1, \alpha_2), (\alpha'_1, \alpha'_2)}(\omega) \\ &= \sum_{v_1, v_2, v'_1, v'_2} u_{v_1, \alpha_1} u_{v_2, \alpha_2}^* G_{(v_1, v_2), (v'_1, v'_2)}(\omega) u_{v'_1, \alpha'_1}^* u_{v'_2, \alpha'_2} . \end{aligned} \quad (114)$$

Here we have added an increment $i\delta$ with $\delta = 0^+$ in order to ensure the correct boundary conditions of a retarded Green's function. For $\tilde{U} = 0$, the inverse Green's function (113) reads

$$\tilde{\Gamma}^{-1}(\omega) \equiv \pm (\omega + i\delta - \tilde{E}) \quad (115)$$

which leads to

$$\Gamma_{A,A'}(\omega) = \Gamma_{(\alpha_1, \alpha_2), (\alpha'_1, \alpha'_2)}(\omega) = \delta_{\alpha_1, \alpha'_1} \delta_{\alpha_2, \alpha'_2} \frac{\rho_{\alpha_2, \alpha_2}^0 - \rho_{\alpha_1, \alpha_1}^0}{\omega - (E_{\alpha_1} - E_{\alpha_2}) + i\delta} . \quad (116)$$

Note that $\tilde{\Gamma}$ is not the exact Green's function for the single-particle Hamiltonian \hat{H}_0 since we just set $\tilde{U} = 0$ in (113), but kept finite the 'Hartree-Fock self-energy' contributions

$$\Sigma_A \equiv \sum_{A'} W_{A,A'} \rho_{A'}^0 \quad (117)$$

which usually change the 'eigenvalues' E_α in (116); c.f. Eqs. (75) and (78).

With the Green's function (116) we can write (113) as

$$\tilde{G}(\omega) = \tilde{\Gamma}(\omega) [1 + \tilde{U} \tilde{\Gamma}(\omega)]^{-1} \quad (118)$$

$$= \tilde{\Gamma}(\omega) + \tilde{\Gamma}(\omega) \tilde{U} \tilde{G}(\omega) \quad (119)$$

where, in the second line, we expanded $[1 + \tilde{U} \tilde{\Gamma}(\omega)]^{-1}$ into a power series with respect to $\tilde{U} \tilde{\Gamma}$. Both Eqs. (118), (119) are familiar expressions for the two-particle Green's function in the random-phase approximation.

5. Time-Dependent Gutzwiller Theory

The time-dependent Gutzwiller approximation has been first introduced for single-band Hubbard models by Seibold et al. [31, 32]. In this section, we generalise this approach for the investigation of multi-band models. To this end, we set up an effective energy functional of the density matrix in section 5.1, which is used in sections 5.2-5.4 to derive the Gutzwiller RPA equations.

5.1. Effective Energy Functional

As summarised in chapter 3, the expectation value of the multi-band Hamiltonian (1) in the Gutzwiller theory is a function of the variational parameters $\lambda_{\Gamma, \Gamma'}$ and of the one-particle wave function $|\Psi_0\rangle$. Like in the Hartree-Fock theory, the single-particle wave function $|\Psi_0\rangle$ enters the energy functional solely through the elements (65) of the non-interacting density matrix $\tilde{\rho}$. It is therefore possible to consider the energy

$$E = E(\vec{\lambda}, \tilde{\rho}) \quad (120)$$

as a function of the density matrix $\tilde{\rho}$ and of the ‘vector’

$$\vec{\lambda} = (\{\lambda_{\Gamma, \Gamma'}^*\}, \{\lambda_{\Gamma, \Gamma'}\}) = (\lambda_1, \dots, \lambda_{n_p}) \quad (121)$$

of n_p variational parameters $\lambda_{\Gamma, \Gamma'}$ (and $\lambda_{\Gamma, \Gamma'}^*$ for $\Gamma \neq \Gamma'$). The density matrix in the energy functional (120) must be derived from a single-particle wave function and, therefore, it has to obey the condition (72). Note that, in the following considerations, the density matrix will either be considered as a matrix (with respect to its two indices (i, σ) and (j, σ')) or as a vector (with respect to its single index Y). To distinguish both cases, we will denote the density matrix $\tilde{\rho}$ in some equations as $\vec{\rho}$ in order to indicate its vector interpretation.

The constraints (35)-(36) are also functions of $\vec{\lambda}$ and $\vec{\rho}$ and will be denoted as

$$g_n(\vec{\lambda}, \vec{\rho}) = 0 \quad , \quad 1 \leq n \leq n_c . \quad (122)$$

Here, n_c is the (maximum) number of independent constraints, which, due to symmetries, is usually smaller than its maximum value $N_{\text{so}}^2 + 1$, where N_{so} is the number of spin-orbital states per lattice site. We assume that the functions (122) are real, i.e., in case of complex equations (33)-(34) their real and imaginary parts are treated separately.

By solving Eqs. (122) we can, at least in principle, express n_c of the variational parameters ($\equiv \lambda_X^d$) through the density matrix ρ_Y and the remaining ‘independent’ parameters ($\equiv \lambda_Z^i$),

$$\lambda_X^d = \lambda_X^d(\vec{\lambda}^i, \vec{\rho}) . \quad (123)$$

In this way, we obtain an energy functional

$$E^{\text{GA}}(\vec{\lambda}^i, \vec{\rho}) \equiv E(\vec{\lambda}^d(\vec{\lambda}^i, \vec{\rho}), \vec{\lambda}^i, \vec{\rho}) . \quad (124)$$

which has to be minimised without constraints apart from Eq. (72) and the condition that the total particle number

$$N = \sum_v \rho_{v,v} \quad (125)$$

is conserved.

For a fixed density matrix $\tilde{\rho}$, the minimisation of (124) with respect to the parameters λ_Z^i ,

$$\frac{\partial}{\partial \lambda_Z^i} E^{\text{GA}}(\vec{\lambda}^i, \vec{\rho}) = 0, \quad (126)$$

determines these parameters

$$\vec{\lambda}^i = \vec{\lambda}^i(\vec{\rho}) \quad (127)$$

as a function of $\vec{\rho}$. This allows us to define the ‘effective’ energy functional

$$E^{\text{eff}}(\vec{\rho}) = E^{\text{GA}}(\vec{\lambda}^i(\vec{\rho}), \vec{\rho}) \quad (128)$$

which, for a fixed density matrix $\vec{\rho}$, is given as the minimum of E^{GA} with respect to $\vec{\lambda}^i$. With this effective functional we will formulate the time-dependent Gutzwiller theory in the following section.

Using a Lagrange-parameter matrix $\tilde{\eta}$ as in chapter 4.1, we find

$$\frac{\partial}{\partial \rho_{v,v'}} \left[E^{\text{eff}}(\tilde{\rho}) - \text{tr}(\tilde{\eta}(\tilde{\rho}^2 - \tilde{\rho})) \right] \Big|_{\tilde{\rho}=\tilde{\rho}^0} = 0 \quad (129)$$

which leads to

$$0 = [\tilde{h}(\tilde{\rho}), \tilde{\rho}]. \quad (130)$$

Here we introduced the matrix $\tilde{h}(\tilde{\rho})$ with the elements

$$h_Y(\tilde{\rho}) = \frac{\partial E^{\text{eff}}(\tilde{\rho})}{\partial \rho_Y}. \quad (131)$$

and used again the notation $\bar{Y} \equiv (j, \sigma'; i, \sigma)$ for $Y = (i, \sigma; j, \sigma')$. The self-consistent solution of Eqs. (130)-(131) then yields the ground-state density matrix $\tilde{\rho}^0$, the matrix $\tilde{h}^0 \equiv \tilde{h}(\tilde{\rho}^0)$, and the corresponding single-particle ‘Gutzwiller-Hamiltonian’

$$\hat{h}^0 \equiv \sum_{i,j;\sigma,\sigma'} h_{i,\sigma;j,\sigma'}^0 \hat{c}_{i,\sigma}^\dagger \hat{c}_{j,\sigma'}. \quad (132)$$

5.2. Gutzwiller RPA Equations

The derivation of RPA-type equations within the time-dependent Gutzwiller theory goes along the same lines as discussed in chapter 4 for the time-dependent Hartree-Fock theory. We add a small time-dependent field

$$\delta \hat{V}_f(t) = \sum_{i,j;\sigma,\sigma'} \delta f_{i,\sigma;j,\sigma'}^0(t) \hat{c}_{i,\sigma}^\dagger \hat{c}_{j,\sigma'} + \text{h.c.} \quad (133)$$

to our multi-band Hamiltonian (1). With the particular time dependence

$$\delta f_{i,\sigma;j,\sigma'}^0(t) = \delta \tilde{f}_{i,\sigma;j,\sigma'}^0(\omega) e^{-i\omega t} \quad (134)$$

the expectation value of $\delta\hat{V}(t)$ reads

$$E_f(\tilde{\rho}) = \sum_{i,j;\sigma,\sigma'} \delta\tilde{f}_{i,\sigma;j,\sigma'}(\omega) e^{-i\omega t} \rho_{j,\sigma';i,\sigma} + \text{c.c.} , \quad (135)$$

where

$$\begin{aligned} \delta\tilde{f}_{i,\sigma_1;j,\sigma_2}(\omega) &= \delta_{i,j} \delta\tilde{f}_{i,\sigma_1;i,\sigma_2}^0(\omega) \frac{C_{i,\sigma_1;i,\sigma_2}^c}{\rho_{i,\sigma_2;i,\sigma_1}} \\ &\quad + (1 - \delta_{i,j}) \sum_{\sigma'_1,\sigma'_2} \delta f_{i,\sigma'_1;j,\sigma'_2}^0(\omega) q_{\sigma'_1}^{\sigma_1} \left(q_{\sigma'_2}^{\sigma_2} \right)^* . \end{aligned} \quad (136)$$

The renormalisation matrix \tilde{q} and the (correlated) local density matrix \tilde{C}^c are defined in equations (27) and (25), respectively. With Eq. (127) they can both be considered as functions of $\tilde{\rho}$.

The time-dependent field induces small fluctuations of the density matrix,

$$\rho_Y = \rho_Y^0 + \delta\rho_Y(t) . \quad (137)$$

Our main assumption is now that $\delta\rho_Y(t)$ obeys the same equation of motion,

$$i\delta\dot{\tilde{\rho}}(t) = [\tilde{h}^0, \delta\tilde{\rho}(t)] + [\delta\tilde{h}(t) + \delta\tilde{f}(t), \tilde{\rho}^0] , \quad (138)$$

as the density matrix in the time-dependent Hartree-Fock theory; see Eq. (98). Here, however, the Hamilton matrix

$$\tilde{h}(t) \approx \tilde{h}^0(t) + \delta\tilde{h}(t) \quad (139)$$

is not derived from the Hartree-Fock functional (68), but from the effective energy functional (128),

$$h_Y(t) = \frac{\partial}{\partial \rho_Y} E^{\text{eff}}(\tilde{\rho}) \approx h_Y^0 + \sum_{Y'} K_{Y,Y'} \delta\rho_{Y'}(t) \equiv h_Y^0 + \delta h_Y(t) , \quad (140)$$

where the matrix \tilde{K} is given as

$$\tilde{K}_{Y,Y'} \equiv \left. \frac{\partial^2 E^{\text{eff}}}{\partial \rho_Y \partial \rho_{Y'}} \right|_{\tilde{\rho}=\tilde{\rho}^0} . \quad (141)$$

The diagonalisation of \tilde{h}^0 (or equivalently of the Gutzwiller Hamiltonian \hat{h}^0) yields a basis $|\alpha\rangle$ with

$$h_{\alpha,\alpha'}^0 = h_A^0 = \delta_{\alpha,\alpha'} E_\alpha \quad (142)$$

and a ground-state density matrix that is given as

$$\rho_{\alpha,\alpha'}^0 = \rho_A^0 = \delta_{\alpha,\alpha'} \Theta(E_F - E_\alpha) . \quad (143)$$

With the projectors $\tilde{\rho}_h \equiv \tilde{\rho}^0$ and $\tilde{\rho}_p \equiv 1 - \tilde{\rho}^0$, we define the particle and hole components of all matrices, as we did in Eqs. (102)-(104). The components $\delta\tilde{\rho}^{vw}(t)$ of the density-matrix fluctuations obey Eqs. (106)-(107), i.e., to leading order we can neglect $\delta\tilde{\rho}^{\text{hh}}(t)$ and $\delta\tilde{\rho}^{\text{pp}}(t)$. Hence, after a Fourier transformation we end up with the same form of RPA equations,

$$\left[(\omega - \tilde{E}) \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix} + \tilde{K} \right] \begin{pmatrix} \delta\tilde{\rho}^{\text{ph}}(\omega) \\ \delta\tilde{\rho}^{\text{hp}}(\omega) \end{pmatrix} = \begin{pmatrix} \delta\tilde{f}^{\text{ph}}(\omega) \\ \delta\tilde{f}^{\text{hp}}(\omega) \end{pmatrix} \quad (144)$$

as in Eq. (111). Here, however, the bare matrix of Coulomb parameters \tilde{U} is replaced by the matrix \tilde{K} , defined in (141), and the energies E_α in the matrix \tilde{E} , Eq. (112), are the eigenvalues of the Gutzwiller Hamiltonian (132). The comparison with (85) leads to the final result

$$\tilde{G}(\omega) \equiv \left[(\omega + i\delta - \tilde{E}) \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix} + \tilde{K} \right]^{-1}. \quad (145)$$

for the two-particle Green's function matrix within the time-dependent Gutzwiller approximation.

One should keep in mind that the external 'fields' $\delta\tilde{f}(\omega)$ in (144) are 'renormalised', i.e., they are not the bare fields as they appear in (85), see Eq. (136). On the other hand, on the l.h.s. of (85) appears the 'correlated' expectation value of the density matrix, while in (144) we work with the fluctuations of the uncorrelated density matrix. Therefore, the 'true' Green's function seen in experiments may, in fact, be given as

$$\underline{G}_{Y,Y'}(\omega) = c_{Y,Y'} G_{Y,Y'}(\omega), \quad (146)$$

with certain frequency independent factors $c_{Y,Y'}$. These factors, however, are of minor importance since they only affect the overall spectral weight and not the frequency dependence of the Green's function matrix $\tilde{G}(\omega)$. We can calculate them with the assumption that correlated and uncorrelated density-matrix fluctuations are related through the same renormalisation factors as the corresponding ground-state density matrices. For the one-band model it has been checked that this prescription is in fact the correct procedure for which the correlation functions fulfil the standard sum rules [34, 37, 43].

5.3. Second-Order Expansion of the Energy Functional

For an evaluation of the Gutzwiller RPA equations (144), we need to determine the matrix \tilde{K} which is given by the second derivatives (141) of the effective energy functional (128). To this end, we expand E^{GA} up to second order around the ground state values $\tilde{\rho}^0$ and $\tilde{\lambda}^{i;0} \equiv \tilde{\lambda}^i(\tilde{\rho}^0)$,

$$\begin{aligned} E^{\text{GA}}(\tilde{\lambda}^i, \tilde{\rho}) &= E_0 + \text{tr}(\tilde{h}^0 \delta\tilde{\rho}) + \frac{1}{2} \left[\sum_{Y,Y'} \delta\rho_Y M_{Y,Y'}^{\rho\rho} \delta\rho_{Y'} + \sum_{Z,Z'} \delta\lambda_Z^i M_{Z,Z'}^{\lambda\lambda} \delta\lambda_{Z'}^i \right. \\ &\quad \left. + \sum_{Z,Y} \left(\delta\lambda_Z^i M_{Z,Y}^{\lambda\rho} \delta\rho_Y + \delta\rho_Y M_{Y,Z}^{\rho\lambda} \delta\lambda_Z^i \right) \right] \\ &\equiv E_0 + \text{tr}(\tilde{h}^0 \delta\tilde{\rho}) + \delta E^{(2)}. \end{aligned} \quad (147)$$

Here, we introduced the matrices $\tilde{M}^{\rho\rho}$, $\tilde{M}^{\lambda\rho}$, $\tilde{M}^{\rho\lambda}$, $\tilde{M}^{\lambda\lambda}$ with the elements

$$M_{Y,Y'}^{\rho\rho} = \frac{\partial^2 E^{\text{GA}}}{\partial\rho_Y \partial\rho_{Y'}}, \quad (148)$$

$$M_{Z,Y}^{\lambda\rho} = \frac{\partial^2 E^{\text{GA}}}{\partial\lambda_Z^i \partial\rho_Y} = M_{Y,Z}^{\rho\lambda}, \quad (149)$$

$$M_{Z,Z'}^{\lambda\lambda} = \frac{\partial^2 E^{\text{GA}}}{\partial\lambda_Z^i \partial\lambda_{Z'}^i}, \quad (150)$$

where the second derivatives on the r.h.s. are evaluated for $\tilde{\rho} = \tilde{\rho}^0$ and $\vec{\lambda}^i = \vec{\lambda}^{i;0}$. Note that there is no linear term $\sim \lambda_Z^i$ in (147) because of the minimisation condition (126). For our further evaluation, it is useful to write the second order terms in Eq. (147) in a more compact form by means of matrix-vector products,

$$\delta E^{(2)} = \frac{1}{2} \left[(\delta \vec{\rho})^T \tilde{M}^{\rho\rho} \delta \vec{\rho} + 2(\delta \vec{\lambda}^i)^T \tilde{M}^{\lambda\rho} \delta \vec{\rho} + (\delta \vec{\lambda}^i)^T \tilde{M}^{\lambda\lambda} \delta \vec{\lambda}^i \right]. \quad (151)$$

Here we used the symmetry

$$\tilde{M}^{\lambda\rho} = [\tilde{M}^{\rho\lambda}]^T. \quad (152)$$

In the effective energy functional (128) the parameters $\vec{\lambda}^i$ are determined by the minimisation condition (126). Applied to our second-order expansion (151) this condition yields

$$\frac{\partial}{\partial \delta \lambda_Z^i} \delta E^{(2)}(\delta \vec{\lambda}^i, \delta \vec{\rho}) = 0, \quad (153)$$

which gives us the multiplet-amplitudes

$$\delta \vec{\lambda}^i = - \left[\tilde{M}^{\lambda\lambda} \right]^{-1} \tilde{M}^{\lambda\rho} \delta \vec{\rho} \quad (154)$$

as a linear function of the densities $\delta \vec{\rho}$. This result leads to the quadratic expansion

$$E^{\text{eff}}(\vec{\rho}^0 + \delta \vec{\rho}) = E_0 + \text{tr}(\tilde{h}^0 \delta \vec{\rho}) + \frac{1}{2} (\delta \vec{\rho})^T \tilde{K} \delta \vec{\rho}, \quad (155)$$

$$\tilde{K} \equiv \tilde{M}^{\rho\rho} - \tilde{M}^{\rho\lambda} \left[\tilde{M}^{\lambda\lambda} \right]^{-1} \tilde{M}^{\lambda\rho}, \quad (156)$$

of the effective energy as a function of the density fluctuations $\delta \vec{\rho}$. In earlier work on the time-dependent Gutzwiller theory, Eqs. (153) and (154) have been denoted as the ‘antiadiabaticity assumption’. In fact, these equations have the physical meaning that the local multiplet dynamics, described by fluctuations $\delta \lambda_Z^i(t)$, are fast compared to those of the density-matrix fluctuations $\delta \rho_Y(t)$. We will use the phrase ‘antiadiabaticity assumption’ in this work too although, strictly speaking, in our derivation it does not constitute an additional approximation.

With the functional (155), we could now proceed with our evaluation of the Gutzwiller RPA Eqs. (144). For practical applications, however, it is more convenient to determine the ‘interaction kernel’ (156) in a way that avoids the explicit solution of the constraint equations (122). This alternative procedure is the subject of the following section.

5.4. Lagrange-functional expansion

In the second-order expansion, described in section 5.3, we implemented the constraints (122) by explicitly eliminating a certain set of n_c variational parameters. Although such a procedure can, at least in principle, always be applied, for the numerical implementation it is more convenient to impose the constraints by means of Lagrange parameters. To this end, we define the ‘Lagrange functional’

$$L(\vec{\lambda}, \vec{\rho}, \vec{\Lambda}) \equiv E(\vec{\lambda}, \vec{\rho}) + \sum_{n=1}^{n_c} \Lambda_n g_n(\vec{\lambda}, \vec{\rho}) \quad (157)$$

which depends on *all* variational parameters $\vec{\lambda}$, the density matrix $\tilde{\rho}(\hat{=}\vec{\rho})$ and the n_c Lagrange parameters Λ_n . The optimum variational parameters λ_Z^0 , density-matrix elements ρ_Y^0 , and Lagrange parameters Λ_n^0 are then determined by the equations

$$\left. \frac{\partial L}{\partial \lambda_Z} \right|_{\vec{\lambda}=\vec{\lambda}^0, \vec{\Lambda}=\vec{\Lambda}^0, \vec{\rho}=\vec{\rho}^0} = \left. \frac{\partial L}{\partial \Lambda_n} \right|_{\dots} = \left. \frac{\partial L}{\partial \rho_Y} \right|_{\dots} = 0 . \quad (158)$$

which have to be solved simultaneously.

We expand the Lagrange functional to leading order with respect to parameter $(\delta\lambda_Z, \delta\Lambda_n)$ and density fluctuations $(\delta\rho_Y)$. The second-order contribution has the form

$$\begin{aligned} \delta L^{(2)} = & \frac{1}{2} \sum_{Y,Y'} \delta\rho_Y L_{Y,Y'}^{\rho\rho} \delta\rho_{Y'} + \sum_{Z,Y} \delta\lambda_Z L_{Z,Y}^{\lambda\rho} \delta\rho_Y + \frac{1}{2} \sum_{Z,Z'} \delta\lambda_Z L_{Z,Z'}^{\lambda\lambda} \delta\lambda_{Z'} \\ & + \sum_n \delta\Lambda_n \left\{ \sum_Z \frac{\partial g_n}{\partial \lambda_Z} \delta\lambda_Z + \sum_Y \frac{\partial g_n}{\partial \rho_Y} \delta\rho_Y \right\} \end{aligned} \quad (159)$$

with matrices $\tilde{L}^{\rho\rho}, \tilde{L}^{\lambda\rho}, \tilde{L}^{\lambda\lambda}$ defined as in Eqs. (148)-(150) only with E^{GA} replaced by L . The antiadiabaticity conditions

$$\frac{\partial}{\partial \delta\lambda_Z} \delta L^{(2)} = 0 , \quad (160)$$

$$\frac{\partial}{\partial \delta\Lambda_n} \delta L^{(2)} = 0 , \quad (161)$$

yield the n_c equations

$$\sum_Z \frac{\partial g_n}{\partial \lambda_Z} \delta\lambda_Z + \sum_Y \frac{\partial g_n}{\partial \rho_Y} \delta\rho_Y = 0 , \quad (162)$$

and the n_p equations

$$\sum_{Z'} L_{Z,Z'}^{\lambda\lambda} \delta\lambda_{Z'} + \sum_Y L_{Z,Y}^{\lambda\rho} \delta\rho_Y + \sum_n \frac{\partial g_n}{\partial \lambda_Z} \delta\Lambda_n = 0 . \quad (163)$$

Together these equations allow us to express the $n_p + n_c$ parameter fluctuations $\delta\Lambda_n, \delta\lambda_Z$ in terms of the density fluctuations $\delta\rho_Y$. These can be reinserted into (159) to obtain the desired quadratic functional solely of the density fluctuations,

$$\delta L^{(2)} = \frac{1}{2} \sum_{Y,Y'} \delta\rho_Y \bar{K}_{Y,Y'} \delta\rho_{Y'} . \quad (164)$$

In Appendix B.1, we prove that the interaction matrix $\bar{K}_{Y,Y'}$ in (164) is, in fact, identical to $K_{Y,Y'}$ in Eqs. (155)-(156).

6. Two particle response functions for lattice models

In the previous chapter we have developed the general formalism of the time-dependent Gutzwiller theory for the calculation of two-particle Green's functions. We will be more specific in this section and explain in detail how the response functions which are of interest in solid-state physics can be calculated within our approach.

6.1. Two-particle response functions

In solid-state physics one is usually not interested in the full two-particle Greens-function \tilde{G} as it has been defined in (82). The properties, relevant for experiments, are certain linear combinations of elements of \tilde{G} . For our translationally invariant model Hamiltonians (1) these are in particular the two-particle response functions

$$G_{(\sigma_2, \sigma_1), (\sigma_3, \sigma_4)}(\vec{R}_i - \vec{R}_j, t - t') \equiv \langle \langle \hat{c}_{i, \sigma_1}^\dagger(t) \hat{c}_{i, \sigma_2}(t); \hat{c}_{j, \sigma_3}^\dagger(t') \hat{c}_{j, \sigma_4}(t') \rangle \rangle \quad (165)$$

or, more importantly, their Fourier transforms

$$\begin{aligned} G_{(\sigma_2, \sigma_1), (\sigma_3, \sigma_4)}(\vec{q}, \omega) &= \frac{1}{L_s} \int_{-\infty}^{\infty} d\tau e^{i\omega\tau} \sum_{i,j} e^{i(\vec{R}_i - \vec{R}_j) \cdot \vec{q}} G_{(\sigma_2, \sigma_1), (\sigma_3, \sigma_4)}(\vec{R}_i - \vec{R}_j, \tau) \\ &= \frac{1}{L_s} \sum_{k, k'} \langle \langle \hat{c}_{k, \sigma_1}^\dagger \hat{c}_{k+q, \sigma_2}; \hat{c}_{k'+q, \sigma_3}^\dagger \hat{c}_{k', \sigma_4} \rangle \rangle_\omega . \end{aligned} \quad (166)$$

Here, we introduced the fermionic operators

$$\hat{c}_{k, \sigma}^{(\dagger)} = \frac{1}{\sqrt{L_s}} \sum_i e^{\mp i \vec{R}_i \cdot \vec{k}} \hat{c}_{i, \sigma}^{(\dagger)} \quad (167)$$

and the usual notation

$$\langle \langle \hat{O}; \hat{O}' \rangle \rangle_\omega = \int_{-\infty}^{\infty} d\tau \langle \langle \hat{O}(\tau); \hat{O}'(0) \rangle \rangle e^{i\omega\tau} \quad (168)$$

for the Fourier transform of a Green's function with arbitrary operators \hat{O}, \hat{O}' . With the abbreviation $v = (\sigma, \sigma')$ for spin-orbit indices and the operators

$$\hat{A}_v^q \equiv \hat{A}_{\sigma_2, \sigma_1}^q \equiv \frac{1}{\sqrt{L_s}} \sum_k \hat{c}_{k, \sigma_1}^\dagger \hat{c}_{k+q, \sigma_2} \quad (169)$$

we can write (166) as

$$G_{v, v'}(\vec{q}, \omega) = \langle \langle \hat{A}_v^q; (\hat{A}_{v'}^q)^\dagger \rangle \rangle_\omega . \quad (170)$$

The Green's functions (166) are still quite general since they include all possible channels of local coupling $\sigma_1 \leftrightarrow \sigma_2$, $\sigma_3 \leftrightarrow \sigma_4$. In experiments one usually measures response functions which are certain linear combinations,

$$G_e(\vec{q}, \omega) = \sum_{v, v'} \kappa_v G_{v, v'}(\vec{q}, \omega) \kappa_{v'} \quad (171)$$

of some of the Green's functions (166), defined by the matrix $\kappa_v = \kappa_{\sigma, \sigma'}$. For example, the transversal spin-susceptibility $\chi(\vec{q}, \omega)$ is given as

$$\chi(\vec{q}, \omega) = \frac{1}{L_s} \langle \langle \hat{S}_q^+; \hat{S}_{-q}^- \rangle \rangle_\omega \quad (172)$$

where

$$\hat{S}_q^+ = \sum_i e^{-i \vec{R}_i \cdot \vec{q}} \hat{S}_i^+ = \sum_k \sum_b \hat{c}_{k, (b\uparrow)}^\dagger \hat{c}_{k+q, (b\downarrow)} , \quad (173)$$

$$\hat{S}_{-q}^- = \sum_i e^{i \vec{R}_i \cdot \vec{q}} \hat{S}_i^- = \sum_k \sum_b \hat{c}_{k+q, (b\downarrow)}^\dagger \hat{c}_{k, (b\uparrow)} \equiv \left(\hat{S}_q^+ \right)^\dagger , \quad (174)$$

$$\hat{S}_i^+ = \sum_b \hat{c}_{i, (b\uparrow)}^\dagger \hat{c}_{i, (b\downarrow)} , \quad \hat{S}_i^- = \sum_b \hat{c}_{i, (b\downarrow)}^\dagger \hat{c}_{i, (b\uparrow)} , \quad (175)$$

are the usual spin-flip operators and b is an index for the orbitals at each lattice site i . The spin susceptibility of a two-band Hubbard model will be investigated in chapter 7.

6.2. Response functions in the time-dependent Gutzwiller approximation

In order to apply the time-dependent Gutzwiller approximation, as developed in chapter 5, we have to expand the Lagrange functional (157) up to second order with respect to density-matrix ($\delta\tilde{\rho}$) and variational-parameter fluctuations ($\delta\lambda_{\Gamma,\Gamma'}$). This means that we need an expansion of the constraints (35)-(36), of the local energies (9)-(11) and (24), and of the kinetic energy (31)-(32). The second-order expansion of the kinetic energy is more involved than that of the local energies and of the constraints. In the latter there are only contributions from fluctuations at *same* lattice sites while in the kinetic energy local and non-local fluctuations (such as $\delta\langle\hat{c}_{i,\sigma}^\dagger\hat{c}_{j,\sigma'}\rangle_{\Psi_0}$) couple. Nevertheless, the calculation of the second-order Lagrange functional is tedious but otherwise straightforward. We therefore refer to Appendix C where the details of this derivation are presented. As shown in that Appendix, it is useful to introduce the operators

$$\hat{B}_w^q \equiv \hat{B}_{\sigma_1,\sigma_2,\sigma'_1,\sigma'_2}^q \equiv \frac{1}{\sqrt{L_s}} \sum_k \epsilon_k^{\sigma_2,\sigma_1} \hat{c}_{k,\sigma'_2}^\dagger \hat{c}_{k+q,\sigma'_1}, \quad (176)$$

$$\hat{\hat{B}}_w^q \equiv \hat{\hat{B}}_{\sigma_1,\sigma_2,\sigma'_1,\sigma'_2}^q \equiv \frac{1}{\sqrt{L_s}} \sum_k \epsilon_{k+q}^{\sigma_2,\sigma_1} \hat{c}_{k,\sigma'_2}^\dagger \hat{c}_{k+q,\sigma'_1}, \quad (177)$$

and to define the auxiliary Green's function matrix $\tilde{\Pi}(\vec{q}, \omega)$ with the elements

$$\Pi_{\substack{v \\ (w)}, \substack{v' \\ (w')}}(\vec{q}, \omega) \equiv \begin{pmatrix} \langle\langle \hat{A}_v^q; (\hat{A}_{v'}^q)^\dagger \rangle\rangle_\omega & \langle\langle \hat{A}_v^q; (\hat{B}_{w'}^q)^\dagger \rangle\rangle_\omega & \langle\langle \hat{A}_v^q; (\hat{\hat{B}}_{w'}^q)^\dagger \rangle\rangle_\omega \\ \langle\langle \hat{B}_w^q; (\hat{A}_{v'}^q)^\dagger \rangle\rangle_\omega & \langle\langle \hat{B}_w^q; (\hat{B}_{w'}^q)^\dagger \rangle\rangle_\omega & \langle\langle \hat{B}_w^q; (\hat{\hat{B}}_{w'}^q)^\dagger \rangle\rangle_\omega \\ \langle\langle \hat{\hat{B}}_w^q; (\hat{A}_{v'}^q)^\dagger \rangle\rangle_\omega & \langle\langle \hat{\hat{B}}_w^q; (\hat{B}_{w'}^q)^\dagger \rangle\rangle_\omega & \langle\langle \hat{\hat{B}}_w^q; (\hat{\hat{B}}_{w'}^q)^\dagger \rangle\rangle_\omega \end{pmatrix}. \quad (178)$$

We are actually interested only in the first 'element' of this matrix, i.e., the Green's functions (170) since they allow us to determine any response function of the form (171). As shown in Appendix D, however, the time-dependent Gutzwiller approximation leads to the following equation for the entire matrix (178) from which (170) can be extracted,

$$\tilde{\Pi}(\vec{q}, \omega) = (1 + \tilde{\Pi}^0(\vec{q}, \omega) \tilde{V}^q)^{-1} \tilde{\Pi}^0(\vec{q}, \omega). \quad (179)$$

Here, \tilde{V}^q is the effective second-order interaction matrix, introduced in (C.40), and $\tilde{\Pi}^0(\vec{q}, \omega)$ is the Green's function matrix (178) evaluated for the single-particle Gutzwiller Hamiltonian (132). As shown in Refs. [10, 46], this Gutzwiller Hamiltonian $\hat{h}^0 \equiv \hat{H}_0^{\text{eff}}$ for our lattice Hamiltonian (1) has the form

$$\hat{H}_0^{\text{eff}} = \sum_k \sum_{\sigma_1, \sigma_2} (\bar{\epsilon}_k^{\sigma_1, \sigma_2} + \eta_{\sigma_1, \sigma_2}) \hat{c}_{k, \sigma_1}^\dagger \hat{c}_{k, \sigma_2} \equiv \sum_k \sum_\alpha E_{k, \alpha} \hat{h}_{k, \alpha}^\dagger \hat{h}_{k, \alpha} \quad (180)$$

where the Lagrange parameters $\eta_{\sigma_1, \sigma_2}$ are determined by the minimisation of the variational ground-state energy and $\bar{\epsilon}_k^{\sigma_1, \sigma_2}$ is defined as

$$\bar{\epsilon}_k^{\sigma_1, \sigma_2} \equiv \sum_{\sigma'_1, \sigma'_2} q_{\sigma'_1}^{\sigma_1} (q_{\sigma'_2}^{\sigma_2})^* \epsilon_k^{\sigma'_1, \sigma'_2}. \quad (181)$$

The creation and annihilation operators $\hat{h}_{k,\alpha}^{(\dagger)}$ of the effective single-particle Hamiltonian (180) can be written as

$$\hat{h}_{k,\alpha}^\dagger = \sum_{\sigma} u_{\sigma,\alpha}^k \hat{c}_{k,\sigma}^\dagger, \quad (182)$$

$$\hat{h}_{k,\alpha} = \sum_{\sigma} (u_{\sigma,\alpha}^k)^* \hat{c}_{k,\sigma}, \quad (183)$$

where the coefficients $u_{\sigma,\alpha}$ are determined by a diagonalisation of (180). With these eigenstates the calculation of $\tilde{\Pi}^0(\vec{q}, \omega)$ is now a simple task. For example, the first element $\langle\langle \hat{A}_v^q; (\hat{A}_{v'}^q)^\dagger \rangle\rangle_\omega^0$ is given as

$$\begin{aligned} & \langle\langle \hat{A}_{\sigma_1, \sigma_2}^q; (\hat{A}_{\sigma'_1, \sigma'_2}^q)^\dagger \rangle\rangle_\omega^0 \\ &= \frac{1}{L_s} \sum_{k, k'} \sum_{\substack{\alpha_1, \alpha_2 \\ \alpha'_1, \alpha'_2}} \langle\langle \hat{h}_{k, \alpha_2}^\dagger \hat{h}_{k+q, \alpha_1}; \hat{h}_{k'+q, \alpha'_1}^\dagger \hat{h}_{k', \alpha'_2} \rangle\rangle_\omega^0 (u_{\sigma_2, \alpha_2}^k)^* u_{\sigma_1, \alpha_1}^{k+q} (u_{\sigma'_1, \alpha'_1}^{k'+q})^* u_{\sigma'_2, \alpha'_2}^{k'} \\ &= \frac{1}{L_s} \sum_k \sum_{\alpha_1, \alpha_2} \frac{(u_{\sigma_2, \alpha_2}^k)^* u_{\sigma_1, \alpha_1}^{k+q} (u_{\sigma'_1, \alpha'_1}^{k+q})^* u_{\sigma'_2, \alpha'_2}^k}{\omega - (E_{k+q, \alpha_1} - E_{k, \alpha_2}) + i\delta} (n_{k, \alpha_2}^0 - n_{k+q, \alpha_1}^0) \end{aligned} \quad (184)$$

where

$$n_{k,\alpha}^0 = \Theta(E_F - E_{k,\alpha}) \quad (185)$$

is the ground-state distribution function (143). In the same way, we can calculate all other elements of $\tilde{\Pi}^0(\vec{q}, \omega)$. The result is always the same as in (184) only with additional factors $\sim \epsilon_k^{\sigma, \sigma'}$ or $\sim \epsilon_{k+q}^{\sigma, \sigma'}$ due to the definition of the operators (176)-(177). For example, the second element in (178) leads to

$$\begin{aligned} & \langle\langle \hat{A}_{\sigma_1, \sigma_2}^q; (\hat{B}_{\sigma_3, \sigma_4, \sigma'_3, \sigma'_4}^q)^\dagger \rangle\rangle_\omega^0 \\ &= \frac{1}{L_s} \sum_k \sum_{\alpha_1, \alpha_2} \frac{(u_{\sigma_2, \alpha_2}^k)^* u_{\sigma_1, \alpha_1}^{k+q} (u_{\sigma'_3, \alpha'_1}^{k+q})^* u_{\sigma'_4, \alpha'_2}^k}{\omega - (E_{k+q, \alpha_1} - E_{k, \alpha_2}) + i\delta} \epsilon_k^{\sigma_3, \sigma_4} (n_{k, \alpha_2}^0 - n_{k+q, \alpha_1}^0). \end{aligned} \quad (186)$$

To summarise, with Eqs. (179), (184), (186), and the interaction matrix (C.40) we are now in the position to investigate any two-particle response function for our general class of multi-band models (1). As a first example, we study the magnetic susceptibility for a two-band model in the following section.

7. Magnetic susceptibility of a two-band Hubbard model

In this chapter we investigate the magnetic susceptibility of a two-band Hubbard model in three spatial dimensions. The model Hamiltonian and the Gutzwiller wave functions which we use for its investigation are introduced in section 7.1. In section 7.2 we discuss the Green's function matrices that we need to study in order to calculate the magnetic susceptibilities within the RPA and the Gutzwiller-RPA schemes. The numerical results for the two-band model are presented in section 7.3.

#	Atomic eigenstate $ \Gamma\rangle$	Symmetry	energy E_Γ
1	$ \uparrow, \uparrow\rangle$	3A_2	$U' - J$
2	$(\uparrow, \downarrow\rangle + \downarrow, \uparrow\rangle)/\sqrt{2}$	3A_2	$U' - J$
3	$ \downarrow, \downarrow\rangle$	3A_2	$U' - J$
4	$(\uparrow, \downarrow\rangle - \downarrow, \uparrow\rangle)/\sqrt{2}$	1E	$U' + J$
5	$(\uparrow\downarrow, 0\rangle - 0, \uparrow\downarrow\rangle)/\sqrt{2}$	1E	$U - J$
6	$(\uparrow\downarrow, 0\rangle + 0, \uparrow\downarrow\rangle)/\sqrt{2}$	1A_1	$U + J$

Table 1. Two-particle eigenstates with symmetry specifications and energies.

7.1. Model and variational ground state

We investigate a Hubbard model with two degenerate e_g orbitals per site on a cubic lattice. The local Hamiltonian (2) for this system can be written as

$$\begin{aligned} \hat{H}_I^{2b} = & U \sum_e \hat{n}_{e,\uparrow} \hat{n}_{e,\downarrow} + U' \sum_{s,s'} \hat{n}_{1,s} \hat{n}_{2,s'} - J \sum_s \hat{n}_{1,s} \hat{n}_{2,s} \\ & + J \sum_s \hat{c}_{1,s}^\dagger \hat{c}_{2,\bar{s}}^\dagger \hat{c}_{1,\bar{s}} \hat{c}_{2,s} + J \left(\hat{c}_{1,\uparrow}^\dagger \hat{c}_{1,\downarrow}^\dagger \hat{c}_{2,\downarrow} \hat{c}_{2,\uparrow} + \hat{c}_{2,\uparrow}^\dagger \hat{c}_{2,\downarrow}^\dagger \hat{c}_{1,\downarrow} \hat{c}_{1,\uparrow} \right). \end{aligned} \quad (187)$$

Here, $e = 1, 2$ labels the e_g orbitals, $s = \uparrow, \downarrow$ is the spin index and we use the convention $\uparrow \equiv \downarrow$, $\downarrow \equiv \uparrow$. Due to the cubic symmetry the Coulomb parameters U , U' and the exchange parameter J are related to each other through

$$U' = U - 2J. \quad (188)$$

Hence, only two of these three parameters can be chosen independently.

There are four spin-orbital states $\sigma = (e, s)$ per atom, leading to a $2^4 = 16$ -dimensional atomic Hilbert space. All eigenstates $|\Gamma\rangle$ of \hat{H}_I^{2b} with particle numbers $N \neq 2$ are simple Slater determinants of spin-orbital states $|\sigma\rangle$ and their energies are

$$\begin{aligned} E_\Gamma &= 0 & (N = 0, 1), \\ E_\Gamma &= U + 2U' - J & (N = 3), \\ E_\Gamma &= 2U + 4U' - 2J & (N = 4). \end{aligned} \quad (189)$$

The two-particle eigenstates are slightly more complicated because some of them are linear combinations of Slater determinants. We introduce the basis

$$|s, s'\rangle \equiv \hat{c}_{1,s}^\dagger \hat{c}_{2,s'}^\dagger |0\rangle, \quad (190)$$

$$|\uparrow\downarrow, 0\rangle \equiv \hat{c}_{1,\uparrow}^\dagger \hat{c}_{1,\downarrow}^\dagger |0\rangle, \quad (191)$$

$$|0, \uparrow\downarrow\rangle \equiv \hat{c}_{2,\uparrow}^\dagger \hat{c}_{2,\downarrow}^\dagger |0\rangle, \quad (192)$$

of two-particle states, which are used to set up the eigenstates of $\hat{H}_{\text{loc};i}$, see table 7.1. The states of lowest energy are the three triplet states with spin $S = 1$, which belong to the representation A_2 of the cubic point-symmetry group. Finding a high-spin ground state is a simple consequence of Hund's first rule. Higher in energy are the two degenerate singlet states of symmetry E and the non-degenerate singlet state of symmetry A_1 .

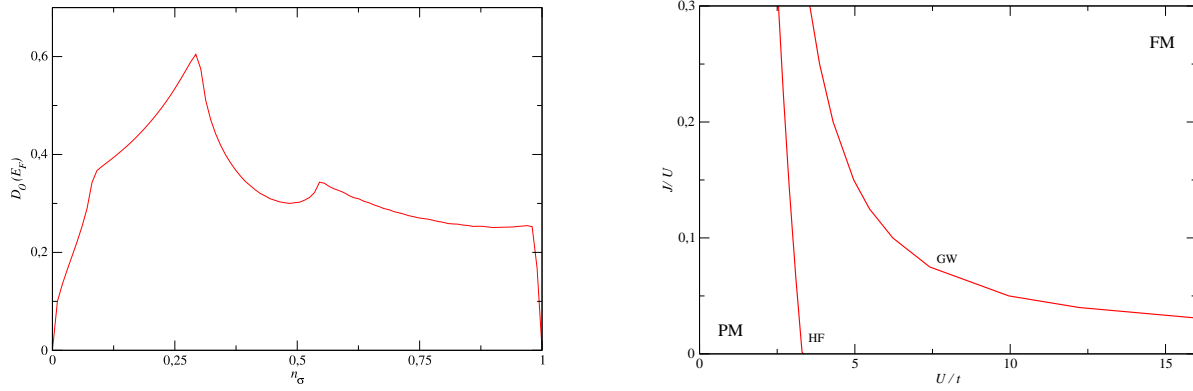


Figure 1. Left: Model density of states at the Fermi energy as a function of orbital filling n_σ . Right: Ground-state phase diagram for both HF and GW. The lines mark the instability for a transition from the paramagnetic (PM) to the ferromagnetic (FM) state. The orbital filling is $n_\sigma \approx 0.29$ and $t \equiv |t_{dd\sigma}^{(1)}|$ (c.f., Ref. [58]).

For the variational ground state we can work with a wave function (4) that contains only diagonal parameters $\lambda_{\Gamma,\Gamma}$. Non-diagonal parameters could only arise if we break the cubic symmetry or want to study states with magnetic orders not collinear to the chosen spin-quantisation axis. Note, however, that for the study of spin excitations we *must* allow for non-diagonal variational parameters, see below.

In our numerical analysis of this two-band model we will consider a tight-binding Hamiltonian \hat{H}_0 with generic hopping parameters which were already used in previous works and lead to the density of states at the Fermi energy shown in Fig. 1 (left). Due to the maximum in the density of states at approx. $n_\sigma = 0.29$, in that range of band fillings there is the strongest tendency for a ferromagnetic state to be lower in energy than the paramagnet. This has already been demonstrated in Ref. [7]. Another important finding in that work is the huge importance of the exchange interaction J for the appearance of ferromagnetic order. This can be seen from the Gutzwiller phase diagram for our model in Fig. 1 (right). In contrast, the HF phase diagrams shows almost no dependence on the size of J ; see also Ref. [44] where similar results have been reported for a two-band model in infinite dimensions.

7.2. The magnetic susceptibility

For the calculation of the spin susceptibility (172), we need to determine a Green's function matrix of the form (178) in which the operators \hat{A}_v^q , \hat{B}_w^q , $\hat{\tilde{B}}_w^q$ are given as

$$\hat{A}_{b_1,b_2}^q \equiv \frac{1}{\sqrt{L_s}} \sum_k \hat{c}_{k,(b_2\uparrow)}^\dagger \hat{c}_{k+q,(b_1\downarrow)} , \quad (193)$$

$$\hat{B}_{b_1,b_2,b'_1,b'_2}^q \equiv \frac{1}{\sqrt{L_s}} \sum_k \epsilon_k^{b_2,b_1} \hat{c}_{k,(b'_2\uparrow)}^\dagger \hat{c}_{k+q,(b'_1\downarrow)} , \quad (194)$$

$$\hat{\tilde{B}}_{b_1,b_2,b'_1,b'_2}^q \equiv \frac{1}{\sqrt{L_s}} \sum_k \epsilon_{k+q}^{b_2,b_1} \hat{c}_{k,(b'_2\uparrow)}^\dagger \hat{c}_{k+q,(b'_1\downarrow)} . \quad (195)$$

The matrix (178), which results from these operators is $4 + 16 + 16 = 36$ dimensional. Due to symmetries, this dimension can be reduced to 20 for a general wave-vector \vec{q} . Along symmetry lines the symmetry reduction could even go further. In our numerical calculations, however, we did not exploit such symmetry considerations since the numerical efforts for a two-band model are still moderate, even in three dimensions.

Note that there is a difference between Hartree-Fock and Gutzwiller RPA calculations concerning the elements of $\tilde{\Pi}^0(\vec{q}, \omega)$ which have to be taken into account in our calculation of the susceptibility

$$\chi(\vec{q}, \omega) = \sum_{b, b'} \langle \langle \hat{A}_{b, b}^q; (\hat{A}_{b', b'}^q)^\dagger \rangle \rangle_\omega. \quad (196)$$

In Hartree-Fock RPA, due to the locality of the interaction terms in Hubbard models and the symmetries of e_g orbitals, the only elements of $\tilde{\Pi}^0(\vec{q}, \omega)$ which contribute are $\langle \langle \hat{A}_{b, b}^q; (\hat{A}_{b', b'}^q)^\dagger \rangle \rangle_\omega^0$, i.e., those which are diagonal with respect to the local orbital indices. This is different from the Gutzwiller RPA equations in which all Green's functions defined by the operators (193)-(195) have to be taken into account. In particular, Green's functions $\langle \langle \hat{A}_{b_1, b_2}^q; (\hat{A}_{b_3, b_4}^q)^\dagger \rangle \rangle_\omega^0$ with $b_1 \neq b_2$ or $b_3 \neq b_4$ cannot be discarded. The reason for this difference is the non-locality of the interaction matrix \tilde{V}^q in the time-dependent Gutzwiller theory.

7.3. Results

We prepare a ferromagnetic ground state in both HF and Gutzwiller approximation at band filling $n_\sigma \approx 0.2987$ in order to be close to the maximum of the DOS. In general both schemes will give different magnetisations for the same set of interaction parameters. Therefore, one could either perform the comparison for fixed parameters or fixed magnetisation (cf. also Ref. [59]). To avoid this inconsistency, we present results for interaction parameters which lead to a fully polarised ferromagnetic ground state in both approximations, i.e. $m = 2n_\sigma$. Note that due to numerical reasons we have to stay slightly below this value in case of the Gutzwiller approximation. The corresponding interaction parameters are specified in the captions to Figs. 2, 3 which display the magnetic excitations obtained within both approximations.

These spectra are composed of a low-energy magnon part due to the breaking of spin-rotational invariance and a high energy Stoner continuum which reflects the particle-hole spin-flip excitations of the 'bare' system, i.e. $\tilde{\Pi}^0(\vec{q}, \omega)$, cf. Eq. (179). For both methods we show the excitations along the (100) and (111) directions. The difference in these directions mainly arises due to the orientation dependence of the particle-hole dispersion which is significantly stronger along the diagonals.

One first important difference between HF and Gutzwiller approximation concerns the difference in the magnetic band splitting $\lambda^- = E_F^\uparrow - E_F^\downarrow$. In HF theory this value is just given by $\lambda^-(HF) = (U + J)m$ and thus for strongly correlated systems produces a large gap $\mathcal{O}(U)$ between the low energy magnon and a high energy Stoner continuum. On the other hand, we find $\lambda^-(GA)$ is significantly reduced with regard to its HF

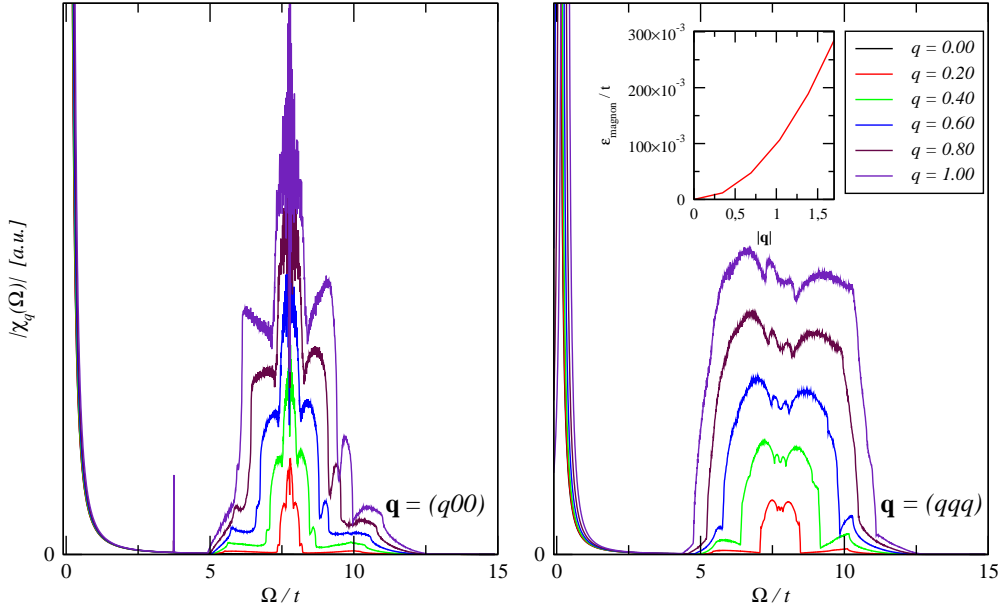


Figure 2. HF excitations for $U = 10.0t$, $J/U = 0.30$ resulting in $m = 0.5975$ (fully polarised) and $\lambda^- = E_F^\uparrow - E_F^\downarrow \approx 7.77t$. The magnon dispersion is fitted by $\varepsilon_{\text{magnon}}(\vec{q})/t = D|\vec{q}|^2(1 + \beta|\vec{q}|^2)$ with $D^{100} \approx D^{111} = 100 \times 10^{-3}$. Scaling: $t \equiv |t_{dd\sigma}^{(1)}|$ (c.f., Ref. [58]) and $q_{x,y,z} \in (-\pi, \pi)$.

counterpart. For the present system $\lambda^-(GA) \approx 1/4\lambda^-(HF)$. Given the broadening of the Stoner continuum with increasing transferred momentum, the low energy magnon thus rapidly merges with the continuum in the time-dependent GA as can be seen from Fig. 3. As a consequence the excitation at $\omega \sim J$, corresponding to a respective spin-flip in the two orbitals, is only visible in HF+RPA along the (100) direction, whereas in the time-dependent Gutzwiller approach it is already within the continuum. Note that the overestimation of the Stoner excitation energy within HF+RPA is a longstanding problem in solid state theory as discussed in Ref. [60].

At $\vec{q} = 0$ all the weight is contained in the zero frequency Goldstone mode. The existence of this excitation provides an important consistency check of the Gutzwiller+RPA approach similar to the analogous finding in HF+RPA. The positive dispersion of the magnon further demonstrates that the underlying Gutzwiller solution is a stable energy minimum which is not destroyed by the fluctuations. The spin-wave stiffness, i.e. the quadratic coefficient of the magnon dispersion, is significantly larger in HF+RPA than in time-dependent Gutzwiller theory. Note, however, that to a certain extend this huge difference is caused by an instability of the ferromagnetic ground state with respect to an incommensurate phase which is found for interaction parameters not much smaller than those used in Fig. 3.

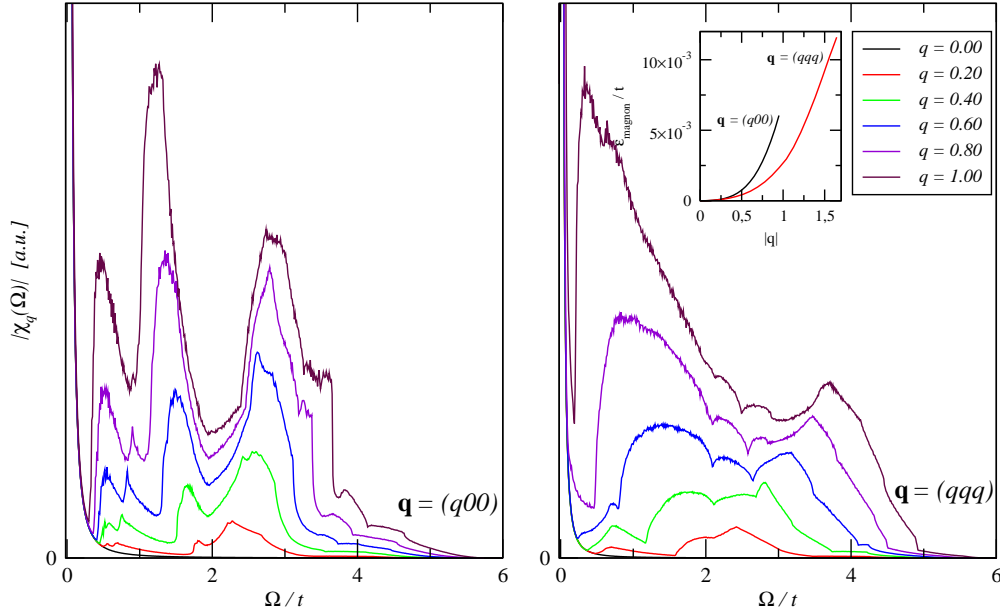


Figure 3. GW excitations for $U = 10.0t$, $J/U = 0.30$ resulting in $m = 0.5728$ (almost fully polarized) and $\lambda^- = E_F^\uparrow - E_F^\downarrow \approx 2.128t$. The Magnon dispersion is fitted by $\varepsilon_{magnon}(\vec{q})/t = D|\vec{q}|^2(1 + \beta|\vec{q}|^2)$ with $D^{100} = 1.34 \times 10^{-3}$ and $D^{111} = 1.30 \times 10^{-3}$ for small wave vectors. Scaling: $t \equiv |t_{dd\sigma}^{(1)}|$ (c.f., Ref. [58]) and $q_{x,y,z} \in (-\pi, \pi)$.

8. Summary

In this paper we have given a detailed derivation of the time-dependent Gutzwiller approximation for multi-band Hubbard models. The basic assumptions which underlie the method can be summarised as follows. First, it is assumed that the dynamics of the Slater-determinant (upon which the Gutzwiller projector acts in the starting Ansatz) is determined by the so-called Gutzwiller Hamiltonian, Eq. (140), which leads to an equation of motion similar to standard RPA, Eq. (138). Second, the dynamics of the variational parameters is determined from the assumption that at each instant of time the energy is minimised. This leads to a linear relation between variational parameter and density fluctuations, Eq. (154). Third, as in the standard HF+RPA approach it is assumed that the external perturbation and thus the density fluctuations are small, Eqs. (106), (107). We have seen that also in the multi-band case these assumptions lead to a consistent theory in the sense, that an instability which is signalled within the Gutzwiller+RPA corresponds to a (second-order) phase transition which one would obtain from the bare variational Gutzwiller approximation. We have further demonstrated that for ferromagnetic ground states the Gutzwiller+RPA leads to the appearance of the Goldstone mode as expected for systems which break continuous spin symmetry.

The formalism as developed in its present form can now be straightforwardly applied to the investigation of correlation functions in strongly correlated multi-band systems as e.g. pnictides, manganites, cobaltates etc. On the other hand, a natural application

of the theory would also comprise the investigation of e.g. orbital quenches for which the small amplitude assumption for the density matrices has to be abandoned. For single-band Hubbard models such a fully time-dependent formulation of the Gutzwiller approximation has been recently presented by Schiró and Fabrizio [61, 62] where also the second assumption above has been replaced by separate equations of motion for the variational parameters. Future work should thus address the question whether their approach reduces to the present theory in the small amplitude limit and how it eventually can be extended to the multi-band case.

Appendix A. Second order expansion of determinants

According to Eq. (16), section 3.1, expectation values $m_{I,I'}^0$ can be written as determinants of certain matrices A with elements that are linear functions of the local density matrix $C_{\gamma,\gamma'}^0$. In the variational ground state, $C_{\gamma,\gamma'}^0$ is diagonal, and, if we chose a proper order of the orbitals γ , to 0-th order the matrix A is diagonal too,

$$m_{I,I'}^0 = |A| = |A^0| = \begin{vmatrix} A_{1,1}^0 & 0 & \dots & 0 \\ 0 & A_{2,2}^0 & \dots & 0 \\ 0 & \dots & \dots & \dots \\ 0 & 0 & 0 & A_{N,N}^0 \end{vmatrix}. \quad (\text{A.1})$$

For $I \neq I'$ at least one of the diagonal elements $A_{i,i}^0$ vanishes and we find

$$m_{I,I'}^0 = \delta_{I,I'} \prod_i A_{i,i} = \delta_{I,I'} \prod_{\gamma \in I} n_\gamma^0 \prod_{\gamma \in (1\dots N) \setminus I} (1 - n_\gamma^0), \quad (\text{A.2})$$

as expected. In order to calculate the first and second derivative of $m_{I,I'}^0$ we need to expand the determinant

$$|A| = |A^0 + \delta a| \quad (\text{A.3})$$

up to second order with respect to the matrix elements $\delta a_{i,j}$. For this expansion one readily finds

$$|A| - |A^0| = |A^0| \sum_i \frac{\delta a_{i,i}}{A_{i,i}^0} + |A^0| \sum_{i,j} \frac{\delta a_{i,i} \delta a_{j,j} + \delta a_{i,j} \delta a_{j,i}}{A_{i,i}^0 A_{j,j}^0}. \quad (\text{A.4})$$

Note that for $A_{i,i}^0 = 0$ the right-hand side is defined by the corresponding limit $A_{i,i}^0 \rightarrow 0$.

Appendix B. Invariance of Second-Order Expansions

Appendix B.1. Equivalence of the Lagrange-functional expansion

In this section, we show that the interaction kernel $\bar{K}_{Y,Y'}^{\rho,\rho'}$ in (164) is identical to $K_{Y,Y'}^{\rho,\rho'}$ in Eqs. (155)-(156). To this end, we choose again some arbitrary independent and dependent variational parameters λ_Z^i and λ_X^d , c.f. Eq. (123). By construction, the constraints (122) are automatically fulfilled as a function of $\vec{\lambda}^i$ and $\vec{\rho}$, i.e., we have

$$g_n(\vec{\lambda}^d(\vec{\lambda}^i, \vec{\rho}), \vec{\lambda}^i, \vec{\rho}) = 0. \quad (\text{B.1})$$

Consequently, all first or higher-order derivatives of (B.1) with respect to λ_Z^i and ρ_Y vanish. For example, the first-order derivatives lead to

$$\frac{dg_n}{d\lambda_Z^i} = \frac{\partial g_n}{\partial \lambda_Z^i} + \sum_X \frac{\partial g_n}{\partial \lambda_X^d} \frac{\partial \lambda_X^d}{\partial \lambda_Z^i} = 0, \quad (\text{B.2})$$

$$\frac{dg_n}{d\rho_Y} = \frac{\partial g_n}{\partial \rho_Y} + \sum_X \frac{\partial g_n}{\partial \lambda_X^d} \frac{\partial \lambda_X^d}{\partial \rho_Y} = 0. \quad (\text{B.3})$$

Using the matrices

$$G_{n,X} \equiv \frac{\partial g_n}{\partial \lambda_X^d}, \quad R_{X,Z} \equiv \frac{\partial \lambda_X^d}{\partial \lambda_Z^i}, \quad Q_{X,Y} \equiv \frac{\partial \lambda_X^d}{\partial \rho_Y}, \quad (\text{B.4})$$

we can write Eqs. (B.2)-(B.3) as

$$\frac{\partial g_n}{\partial \lambda_Z^i} = -[\tilde{G}\tilde{R}]_{n,Z}, \quad (\text{B.5})$$

$$\frac{\partial g_n}{\partial \rho_Y} = -[\tilde{G}\tilde{Q}]_{n,Y}. \quad (\text{B.6})$$

With the classification of dependent and independent variables we are in the position to evaluate the antiadiabaticity conditions (162)-(163). First, Eq. (162) leads to

$$\sum_X \frac{\partial g_n}{\partial \lambda_X^d} \delta \lambda_X^d + \sum_Z \frac{\partial g_n}{\partial \lambda_Z^i} \delta \lambda_Z^i + \sum_Y \frac{\partial g_n}{\partial \rho_Y} \delta \rho_Y = 0 \quad (\text{B.7})$$

which, together with Eqs. (B.5)-(B.6), yields

$$\tilde{G} [\delta \vec{\lambda}^d - \tilde{R} \delta \vec{\lambda}^i - \tilde{Q} \delta \vec{\rho}] = \vec{0}. \quad (\text{B.8})$$

Since the square matrix \tilde{G} should be invertible, the bracket in (B.8) must vanish. Hence, we find the relation

$$\delta \vec{\lambda}^d = \tilde{R} \delta \vec{\lambda}^i + \tilde{Q} \delta \vec{\rho} \quad (\text{B.9})$$

which determines the dependent-parameters fluctuations $\delta \vec{\lambda}^d$ as a function of $\delta \vec{\lambda}^i$ and $\delta \vec{\rho}$.

Applying the separation of dependent and independent parameter fluctuations to the second set of Eqs. (163) yields

$$\begin{pmatrix} \tilde{A}^T \\ \tilde{G}^T \end{pmatrix} \delta \vec{\Lambda} = - \begin{pmatrix} \tilde{L}^{ii} & \tilde{L}^{id} \\ \tilde{L}^{di} & \tilde{L}^{dd} \end{pmatrix} \begin{pmatrix} \delta \vec{\lambda}^i \\ \delta \vec{\lambda}^d \end{pmatrix} - \begin{pmatrix} \tilde{L}^{i\rho} \\ \tilde{L}^{d\rho} \end{pmatrix} \delta \vec{\rho}. \quad (\text{B.10})$$

Here we introduced the six matrices

$$L_{Z,Z'}^{ii} \equiv \frac{\partial^2 L}{\partial \lambda_Z^i \partial \lambda_{Z'}^i}, \dots, L_{X,Y}^{d\rho} \equiv \frac{\partial^2 L}{\partial \lambda_X^d \partial \rho_Y}, \quad (\text{B.11})$$

of second derivatives. With (B.9) and the second ‘row’ of Eqs. (B.10) one can write the Lagrange-parameter fluctuations as a function of $\delta \vec{\lambda}^i$ and $\delta \vec{\rho}$,

$$\delta \vec{\Lambda} = -[\tilde{G}^T]^{-1} \left[(\tilde{L}^{di} + \tilde{L}^{dd} \tilde{R}) \delta \vec{\lambda}^i + (\tilde{L}^{dd} \tilde{Q} + \tilde{L}^{d\rho}) \delta \vec{\rho} \right]. \quad (\text{B.12})$$

Inserting this expression into the first row of Eqs. (B.10) and using

$$\tilde{A}^T = -\tilde{R}^T \tilde{G}^T \quad (\text{B.13})$$

we eventually find

$$\begin{aligned} \delta \vec{\lambda}^i = & - \left[\tilde{L}^{ii} + \tilde{L}^{id} \tilde{R} + \tilde{R}^T + \tilde{R}^T \tilde{L}^{dd} \tilde{R} \right]^{-1} \\ & \times \left[\tilde{L}^{ip} + \tilde{L}^{id} \tilde{Q} + \tilde{R}^T \tilde{L}^{dp} + \tilde{R}^T \tilde{L}^{dd} \tilde{Q} \right] \delta \vec{\rho}. \end{aligned} \quad (\text{B.14})$$

Equations (B.14), (B.12), and (B.9) now enable us to write all fluctuations $\delta \vec{\lambda}^i$, $\delta \vec{\lambda}^d$, and $\delta \vec{\Lambda}$ as functions of the density fluctuations $\delta \vec{\rho}$. These relations can be inserted into the second-order expansion of the Lagrange functional,

$$\begin{aligned} 2\delta L^{(2)} = & (\delta \vec{\rho})^T \tilde{L}^{\rho\rho} \delta \vec{\rho} + (\delta \vec{\lambda}^i)^T \tilde{L}^{ii} \delta \vec{\lambda}^i + (\delta \vec{\lambda}^d)^T \tilde{L}^{dd} \delta \vec{\lambda}^d \\ & + \left[((\delta \vec{\rho})^T \tilde{L}^{\rho d} \delta \vec{\lambda}^d + (\delta \vec{\rho})^T \tilde{L}^{\rho i} \delta \vec{\lambda}^i + (\delta \vec{\lambda}^i)^T \tilde{L}^{id} \delta \vec{\lambda}^d) \right. \\ & \left. + (\dots)^T \right] + 2(\delta \vec{\Lambda})^T \tilde{G} \left[\delta \vec{\lambda}^d - \tilde{R} \delta \vec{\lambda}^i - \tilde{Q} \delta \vec{\rho} \right] \end{aligned} \quad (\text{B.15})$$

in order to calculate $\bar{K}_{Y,Y'}^{\rho\rho}$ in Eq. (164). However, to prove just the identity of $\bar{K}_{Y,Y'}^{\rho\rho}$, and $K_{Y,Y'}^{\rho\rho}$ in (155) it is sufficient to apply only Eq. (B.9) to the expansion (B.15). This leads to

$$\begin{aligned} 2\delta L^{(2)} = & (\delta \vec{\rho})^T (\tilde{L}^{\rho\rho} + \tilde{Q}^T \tilde{L}^{dp} + \tilde{L}^{\rho d} \tilde{Q} + \tilde{Q}^T \tilde{L}^{dd} \tilde{Q}) \delta \vec{\rho} \\ & + (\delta \vec{\lambda}^i)^T (\tilde{L}^{ii} + \tilde{L}^{id} \tilde{R} + \tilde{R}^T \tilde{L}^{di} + \tilde{R}^T \tilde{L}^{dd} \tilde{R}) \delta \vec{\lambda}^i \\ & + \left[(\delta \vec{\rho})^T (\tilde{L}^{\rho i} + \tilde{L}^{\rho i} \tilde{R} + \tilde{Q}^T \tilde{L}^{di} + \tilde{Q}^T \tilde{L}^{dd} \tilde{R}) \delta \vec{\lambda}^i + (\dots)^T \right]. \end{aligned} \quad (\text{B.16})$$

As we will show below, the matrices (148)-(150) which determine the second order expansion (151) are the same as the corresponding matrices in (B.16). Hence, we have

$$\delta E^{(2)} = \delta L^{(2)}. \quad (\text{B.17})$$

Since the antiadiabaticity condition

$$\frac{\partial \delta E^{(2)}}{\partial \delta \lambda_Z^i} = \frac{\partial \delta L^{(2)}}{\partial \delta \lambda_Z^i} = 0 \quad (\text{B.18})$$

for $\delta E^{(2)}$ reproduces Eq. (B.14), the identity of $\bar{K}_{Y,Y'}^{\rho\rho}$, and $K_{Y,Y'}^{\rho\rho}$ is then finally demonstrated.

It remains to be shown that the matrices (148)-(150) agree with those in (B.16). To this end, we use the explicit form (124) of the energy functional (120) that appears in the definition of the matrices (148)-(150). As an example, we consider the matrix $\tilde{M}^{\rho\rho}$ and show that it is identical to the matrix in the first line of (B.16). With similar derivations one can prove the same for the other matrices (149),(150) and their counterparts in (B.16).

Using (124) and (148) we find

$$M_{Y,Y'}^{\rho\rho} = [\tilde{E}^{\rho\rho} + \tilde{Q}^T \tilde{E}^{dp} + \tilde{E}^{\rho d} \tilde{Q} + \tilde{Q}^T \tilde{E}^{dd} \tilde{Q}]_{Y,Y'} + 2 \sum_X \frac{\partial E}{\partial \lambda_X^d} \cdot \frac{\partial^2 \lambda_X^d}{\partial \rho_Y \partial \rho_{Y'}}. \quad (\text{B.19})$$

Here, the matrices

$$\tilde{E}^{\rho\rho} = \tilde{L}^{\rho\rho} - \sum_n \Lambda_n \tilde{g}_n^{\rho\rho}, \dots, \tilde{E}^{\text{dd}} = \tilde{L}^{\text{dd}} - \sum_n \Lambda_n \tilde{g}_n^{\text{dd}}, \quad (\text{B.20})$$

and $\tilde{g}_n^{\rho\rho}, \dots, \tilde{g}_n^{\text{dd}}$ are defined as in (B.11) only with L replaced by E or g_n respectively. Obviously, the matrix in the first line of (B.16) is identical to $\tilde{M}^{\rho\rho}$ if

$$2 \sum_X \frac{\partial E}{\partial \lambda_X^{\text{d}}} \cdot \frac{\partial^2 \lambda_X^{\text{d}}}{\partial \rho_Y \partial \rho_{Y'}} = - \sum_n \Lambda_n [\tilde{g}_n^{\rho\rho} + \tilde{Q}^T \tilde{g}_n^{\text{d}\rho} + \tilde{g}_n^{\rho\text{d}} \tilde{Q} + \tilde{Q}^T \tilde{g}_n^{\text{dd}} \tilde{Q}]_{Y,Y'}. \quad (\text{B.21})$$

To prove (B.21), we use the fact that the second (total) derivatives of (B.1) with respect to the densities ρ_Y vanish

$$\begin{aligned} \frac{dg_n}{d\rho_Y d\rho_{Y'}} &= [\tilde{g}_n^{\rho\rho} + \tilde{Q}^T \tilde{g}_n^{\text{d}\rho} + \tilde{g}_n^{\rho\text{d}} \tilde{Q} + \tilde{Q}^T \tilde{g}_n^{\text{dd}} \tilde{Q}]_{Y,Y'} + 2 \sum_X \frac{\partial g_n}{\partial \lambda_X^{\text{d}}} \cdot \frac{\partial^2 \lambda_X^{\text{d}}}{\partial \rho_Y \partial \rho_{Y'}} \\ &= 0. \end{aligned} \quad (\text{B.22})$$

Equation (B.21) is therefore fulfilled if

$$\sum_X \left(\frac{\partial E}{\partial \lambda_X^{\text{d}}} + \sum_n \Lambda_n \frac{\partial g_n}{\partial \lambda_X^{\text{d}}} \right) \frac{\partial^2 \lambda_X^{\text{d}}}{\partial \rho_Y \partial \rho_{Y'}} = 0. \quad (\text{B.23})$$

This equation, however, holds trivially, since (158) leads to

$$\frac{\partial L}{\partial \lambda_Z} = \frac{\partial E}{\partial \lambda_Z} + \sum_n \Lambda_n \frac{\partial g_n}{\partial \lambda_Z} = 0 \quad (\text{B.24})$$

for all parameters λ_Z and in particular for $\lambda_Z = \lambda_X^{\text{d}}$ as it appears in (B.24).

Appendix B.2. Linear transformations of the density matrix

In investigations of our translationally invariant lattice systems (1) it turns out to be more convenient to work with fluctuations $\delta\vec{\mu}$ which are linearly related to the density-matrix fluctuations,

$$\delta\vec{\rho} = \tilde{\Xi} \cdot \delta\vec{\mu} \quad (\text{B.25})$$

c.f., Eqs. (C.27)-(C.28) and the resulting Green's functions (178). The effective second-order functional (155)-(156) in terms of the fluctuations $\delta\vec{\mu}$ is then given as

$$\delta E^{(2)}(\delta\vec{\mu}) = \frac{1}{2} (\delta\vec{\mu})^T (\tilde{\Xi}^T \tilde{K}^{\rho\rho} \tilde{\Xi}) \delta\vec{\mu} \quad (\text{B.26})$$

with $\tilde{K}^{\rho\rho}$ as defined in (156). For numerical calculations it is important to show that one obtains the same kernel

$$\tilde{K}^{\mu\mu} \equiv \tilde{\Xi}^T \tilde{K}^{\rho\rho} \tilde{\Xi} \quad (\text{B.27})$$

as in (B.26) if the transformation (B.25) and the antiadiabaticity condition are applied in the reverse order: If we apply (B.25) first to (151), we obtain

$$\delta E^{(2)} = \frac{1}{2} \left[(\delta\vec{\mu})^T \tilde{\Xi}^T \tilde{M}^{\rho\rho} \tilde{\Xi} \delta\vec{\mu} + 2(\delta\vec{\lambda}^{\text{i}})^T \tilde{M}^{\lambda\rho} \tilde{\Xi} \delta\vec{\mu} + (\delta\vec{\lambda}^{\text{i}})^T \tilde{M}^{\lambda\lambda} \delta\vec{\lambda}^{\text{i}} \right]. \quad (\text{B.28})$$

The antiadiabaticity condition for $\delta\vec{\mu}$ then reads

$$\delta\vec{\lambda}^i = - \left[\tilde{M}^{\lambda\lambda} \right]^{-1} \tilde{M}^{\lambda\rho} \tilde{\Xi} \delta\vec{\mu} . \quad (\text{B.29})$$

Inserted into (B.28) this equation yields

$$\begin{aligned} \delta E^{(2)}(\delta\vec{\mu}) &= E_0 + \frac{1}{2} (\delta\vec{\mu})^T \tilde{K}^{\mu\mu} \delta\vec{\mu} , \\ \tilde{K}^{\mu\mu} &= \tilde{\Xi}^T \tilde{M}^{\rho\rho} \tilde{\Xi} - \tilde{\Xi}^T \tilde{M}^{\rho\lambda} \left[\tilde{M}^{\lambda\lambda} \right]^{-1} \tilde{M}^{\lambda\rho} \tilde{\Xi} = \tilde{\Xi}^T \tilde{K}^{\rho\rho} \tilde{\Xi} , \end{aligned} \quad (\text{B.30})$$

as claimed above.

Appendix C. Explicit form of the second-order expansion

We calculate the second-order expansion of the Lagrange functional with respect to the variational parameters $\lambda_{i;\Gamma,\Gamma'}$ and the density matrix (65). For the general consideration in section 5 and Appendix B it was convenient to subsume the parameters $\lambda_{\Gamma,\Gamma'}$ and their conjugates $\lambda_{\Gamma,\Gamma'}^*$ in a set of n_p parameters λ_Z , c.f., Eq. (121). Here in this appendix, where we aim to resolve the explicit structure of the second-order expansion, it is better to take the difference between $\lambda_{\Gamma,\Gamma'}$ and $\lambda_{\Gamma,\Gamma'}^*$ into account.

The constraints (35)-(36), the local energy (24), and the renormalisation matrix (27) are all functions only of $\lambda_{i;\Gamma,\Gamma'}^*$, $\lambda_{i;\Gamma,\Gamma'}$ and of the local density matrix $C_{i;\sigma,\sigma'}^0$. For simplicity we use the joint variables $A_v^i, (A_v^i)^*$ for all these local variables, i.e., it is either

$$A_v^i = A_{\sigma_1,\sigma_2}^i = \langle \hat{c}_{i,\sigma_2}^\dagger \hat{c}_{i,\sigma_1} \rangle \quad \text{or} \quad A_v^i = A_{\Gamma,\Gamma'}^i = \lambda_{i;\Gamma,\Gamma'} . \quad (\text{C.1})$$

With respect to the parameters $\lambda_{i;\Gamma,\Gamma'}^*$, $\lambda_{i;\Gamma,\Gamma'}$ the second derivatives of (35)-(36), (24), and (27) are quadratic functions of the form $\sim (A_v^i)^* A_{v'}^i$. Due to the Hermiticity of the density matrix the same can be achieved with respect to the local density matrix. Then the only finite second derivatives of the Lagrange functional

$$L = T + \sum_i E_{i,\text{loc}}(\{(A_v^i)^*\}, \{A_v^i\}) + \sum_{i,n} \Lambda_{i,n} g_{i,n}(\{(A_v^i)^*\}, \{A_v^i\}) \quad (\text{C.2})$$

$$\begin{aligned} &\equiv T + L_{\text{loc}} \\ T &= \sum_{i \neq j} \sum_{\substack{\sigma_1, \sigma_2 \\ \sigma_1', \sigma_2'}} t_{i,j}^{\sigma_1, \sigma_2} q_{i,\sigma_1}^{\sigma_1'} \left(q_{j,\sigma_2}^{\sigma_2'} \right)^* \langle \hat{c}_{i\sigma_1'}^\dagger \hat{c}_{j\sigma_2'} \rangle \end{aligned} \quad (\text{C.3})$$

are

$$\frac{\partial^2 L}{\partial (A_v^i)^* \partial A_{v'}^i} \neq 0 \quad (\text{C.4})$$

whereas

$$\frac{\partial^2 L}{\partial (A_v^i)^* \partial (A_{v'}^i)^*} = \frac{\partial^2 L}{\partial A_v^i \partial A_{v'}^i} = 0 . \quad (\text{C.5})$$

The second-order expansion of the constraints and the local energy is straightforward since only local fluctuations δA_v^i couple,

$$\delta L_{\text{loc}}^{(2)} = \sum_q \sum_{v,v'} (\delta A_v^q)^* K_{v,v'}^{\text{loc}} \delta A_{v'}^q \quad (\text{C.6})$$

where we introduced

$$K_{v,v'}^{\text{loc}} = \frac{\partial^2 L_{\text{loc}}}{\partial(A_v^i)^* \partial A_{v'}^i} \quad (\text{C.7})$$

and the Fourier transforms of the local fluctuations

$$\delta A_v^i = \frac{1}{\sqrt{L_s}} \sum_q e^{-i\vec{R}_i \cdot \vec{q}} \delta A_v^q. \quad (\text{C.8})$$

All derivatives in this section (e.g., (C.7)) have to be evaluated for the ground-state values of the variational parameters $\lambda_{i,\Gamma,\Gamma'}$, the density matrix $\tilde{\rho}$, and the Lagrange parameters $\Lambda_{i,n}$. Note that the density-matrix fluctuations $\delta A_{\sigma_2,\sigma_1}^q$ can be written as

$$\begin{aligned} \delta A_{\sigma_2,\sigma_1}^q &= \frac{1}{\sqrt{L_s}} \sum_i e^{i\vec{R}_i \cdot \vec{q}} \delta \langle \hat{c}_{i,\sigma_1}^\dagger \hat{c}_{i,\sigma_2} \rangle = \frac{1}{\sqrt{L_s}} \sum_k \delta \langle \hat{c}_{k,\sigma_1}^\dagger \hat{c}_{k+q,\sigma_2} \rangle \\ &= \delta \langle \hat{A}_{\sigma_2,\sigma_1}^q \rangle \end{aligned} \quad (\text{C.9})$$

where the operator \hat{A}_v^q has been defined in (169).

In addition to (C.6), we need to take into account the mixed terms $\sim \delta A_v^i \delta \Lambda_{i,n}$. In real space, their contribution is given as

$$\delta L_c^{(2)} = \sum_{i,n,v} \left(\frac{\partial g_{i,n}}{\partial(A_v^i)^*} \delta(A_v^i)^* + \frac{\partial g_{i,n}}{\partial A_v^i} \delta A_v^i \right) \delta \Lambda_{i,n}. \quad (\text{C.10})$$

If we introduce the Fourier transforms $\delta \Lambda_n^q$ of the fluctuations $\delta \Lambda_{i,n}$, we can write (C.10) as

$$\delta L_c^{(2)} = \sum_q \sum_{n,v} (\delta A_v^q)^* K_{v,n}^c \delta \Lambda_n^q + (\delta \Lambda_n^q)^* (K_{v,n}^c)^* \delta A_v^q. \quad (\text{C.11})$$

Here, we used that the constraints $g_{i,n}$ are assumed to be real and lattice-site independent such that

$$K_{v,n}^c \equiv \frac{\partial g_{i,n}}{\partial(A_v^i)^*} = \left(\frac{\partial g_{i,n}}{\partial A_v^i} \right)^*. \quad (\text{C.12})$$

More involved than the calculation of (C.6) is the expansion of the kinetic energy. Here we find

$$\delta T^{(2)} = \delta T_1^{(2)} + \delta T_t^{(2)} \quad (\text{C.13})$$

with

$$\begin{aligned} \delta T_1^{(2)} &= \sum_{i \neq j} \sum_{\sigma_1, \sigma_2, \sigma'_1, \sigma'_2} t_{i,j}^{\sigma_1, \sigma_2} \langle \hat{c}_{i,\sigma'_1}^\dagger \hat{c}_{j,\sigma'_2} \rangle \sum_{v,v'} \left[\frac{\partial^2 q_{i,\sigma_1}^{\sigma'_1}}{\partial(A_v^i)^* \partial A_{v'}^i} (q_{j,\sigma_2}^{\sigma'_2})^* (\delta A_v^i)^* \delta A_{v'}^i \right. \\ &\quad \left. + \frac{1}{2} \left(\frac{\partial q_{i,\sigma_1}^{\sigma'_1}}{\partial(A_v^i)^*} \frac{\partial (q_{j,\sigma_2}^{\sigma'_2})^*}{\partial A_{v'}^j} (\delta A_v^i)^* \delta A_{v'}^j + \frac{\partial q_{i,\sigma_1}^{\sigma'_1}}{\partial A_v^i} \frac{\partial (q_{j,\sigma_2}^{\sigma'_2})^*}{\partial(A_{v'}^j)^*} \delta A_v^i (\delta A_{v'}^j)^* \right) \right] \\ &\quad + \text{c.c.} \end{aligned} \quad (\text{C.14})$$

and

$$\begin{aligned} \delta T_t^{(2)} = & \sum_{i \neq j} \sum_{\sigma_1, \sigma_2, \sigma'_1, \sigma'_2} t_{i,j}^{\sigma_1, \sigma_2} \delta \langle \hat{c}_{i\sigma'_1}^\dagger \hat{c}_{j\sigma'_2} \rangle \\ & \times \sum_v \left[\frac{\partial q_{i,\sigma_1}^{\sigma'_1}}{\partial (A_v^i)^*} \left(q_{j,\sigma_2}^{\sigma'_2} \right)^* (\delta A_v^i)^* + q_{i,\sigma_1}^{\sigma'_1} \frac{\partial \left(q_{j,\sigma_2}^{\sigma'_2} \right)^*}{\partial (A_v^j)^*} (\delta A_v^j)^* \right] + \text{c.c.} . \end{aligned} \quad (\text{C.15})$$

The fact that the complex conjugates give the terms not explicitly shown in Eqs. (C.14)-(C.15) follows from the relations

$$\left(\frac{\partial q_{\sigma}^{\sigma'}}{\partial A_v} \right)^* = \frac{\partial \left(q_{\sigma}^{\sigma'} \right)^*}{\partial (A_v)^*} , \quad (\text{C.16})$$

$$\left(\frac{\partial^2 q_{\sigma}^{\sigma'}}{\partial (A_v)^* \partial A_{v'}} \right)^* = \frac{\partial^2 \left(q_{\sigma}^{\sigma'} \right)^*}{\partial (A_{v'})^* \partial A_v} , \quad (\text{C.17})$$

$$\left(t_{i,j}^{\sigma, \sigma'} \right)^* = t_{j,i}^{\sigma', \sigma} . \quad (\text{C.18})$$

For our translationally invariant ground state it is more convenient to write (C.14)-(C.15) in momentum space: With the Fourier transforms of the local fluctuations the term (C.14) reads

$$\delta T_1^{(2)} = \sum_q \sum_{v, v'} (\delta A_v^q)^* [K_{q;v,v'}^1 + (K_{q;v',v}^1)^*] \delta A_{v'}^q \quad (\text{C.19})$$

where

$$\begin{aligned} K_{q;v,v'}^1 \equiv & \sum_{\sigma_1, \sigma_2, \sigma'_1, \sigma'_2} \left[\frac{1}{2} E_{\sigma_1, \sigma_2, \sigma'_1, \sigma'_2}(\vec{q}) \left(\frac{\partial q_{\sigma_1}^{\sigma'_1}}{\partial (A_v)^*} \frac{\partial \left(q_{\sigma_2}^{\sigma'_2} \right)^*}{\partial A_{v'}} + \frac{\partial q_{\sigma_1}^{\sigma'_1}}{\partial A_v} \frac{\partial \left(q_{\sigma_2}^{\sigma'_2} \right)^*}{\partial (A_v)^*} \right) \right. \\ & \left. + E_{\sigma_1, \sigma_2, \sigma'_1, \sigma'_2} \frac{\partial^2 q_{\sigma_1}^{\sigma'_1}}{\partial (A_v)^* \partial A_{v'}} \left(q_{\sigma_2}^{\sigma'_2} \right)^* \right] . \end{aligned} \quad (\text{C.20})$$

Here we assumed that the renormalisation matrix is lattice-site independent and introduced the tensor

$$E_{\sigma_1, \sigma_2, \sigma'_1, \sigma'_2}(\vec{q}) = \frac{1}{L_s} \sum_k \epsilon_{k+q}^{\sigma_1, \sigma_2} \langle \hat{c}_{k\sigma'_1}^\dagger \hat{c}_{k\sigma'_2} \rangle \quad (\text{C.21})$$

with

$$\epsilon_k^{\sigma_1, \sigma_2} = \frac{1}{L_s} \sum_{i \neq j} t_{i,j}^{\sigma_1, \sigma_2} e^{i\vec{k}(\vec{R}_i - \vec{R}_j)} . \quad (\text{C.22})$$

Note that for $\vec{q} = 0$ the tensor (C.21),

$$E_{\sigma_1, \sigma_2, \sigma'_1, \sigma'_2} = E_{\sigma_1, \sigma_2, \sigma'_1, \sigma'_2}(0) \quad (\text{C.23})$$

has already been defined in (32). For the evaluation of the second ('transitive') term (C.15) we write the non-local density-matrix fluctuations as

$$\delta \langle \hat{c}_{i\sigma'_1}^\dagger \hat{c}_{j\sigma'_2} \rangle = \frac{1}{L_s} \sum_{k, k'} e^{i(\vec{R}_i \cdot \vec{k} - \vec{R}_j \cdot \vec{k}')} \delta \langle \hat{c}_{k\sigma'_1}^\dagger \hat{c}_{k'\sigma'_2} \rangle . \quad (\text{C.24})$$

Together with (C.8) this yields

$$\delta T_t^{(2)} = \frac{1}{L_s} \sum_{q,k} \sum_{v;\sigma'_1,\sigma'_2} (\delta A_v^q)^* \bar{K}_{k,q;v,\sigma'_1,\sigma'_2}^t \delta \langle \hat{c}_{k\sigma'_1}^\dagger \hat{c}_{k+q\sigma'_2} \rangle + \text{c.c} \quad (\text{C.25})$$

with

$$\bar{K}_{k,q;v,\sigma'_1,\sigma'_2}^t = \sum_{\sigma_1,\sigma_2} \left[\frac{\partial q_{\sigma_1}^{\sigma'_1}}{\partial (A_v)^*} \left(q_{\sigma_2}^{\sigma'_2} \right)^* \epsilon_{k+q}^{\sigma_1,\sigma_2} + q_{\sigma_1}^{\sigma'_1} \frac{\partial \left(q_{\sigma_2}^{\sigma'_2} \right)^*}{\partial (A_v)^*} \epsilon_k^{\sigma_1,\sigma_2} \right]. \quad (\text{C.26})$$

In principle, Eqs. (C.25)-(C.26) allow us to calculate all second-order couplings of density-matrix and parameter fluctuations that arise from $\delta T_t^{(2)}$. For numerical calculations, however, these equations are not very useful due to the explicit k dependence of (C.26). It is much easier to introduce the two auxiliary fluctuations

$$\delta B_w^q \equiv \delta B_{\sigma_2,\sigma_1,\sigma'_2,\sigma'_1}^q \equiv \frac{1}{\sqrt{L_s}} \sum_k \epsilon_k^{\sigma_1,\sigma_2} \delta \langle \hat{c}_{k\sigma'_1}^\dagger \hat{c}_{k+q\sigma'_2} \rangle, \quad (\text{C.27})$$

$$\delta \bar{B}_w^q \equiv \delta \bar{B}_{\sigma_2,\sigma_1,\sigma'_2,\sigma'_1}^q \equiv \frac{1}{\sqrt{L_s}} \sum_k \epsilon_{k+q}^{\sigma_1,\sigma_2} \delta \langle \hat{c}_{k\sigma'_1}^\dagger \hat{c}_{k+q\sigma'_2} \rangle, \quad (\text{C.28})$$

where $w \equiv (\sigma_2, \sigma_1, \sigma'_2, \sigma'_1)$ is an abbreviation for quadruples of indices σ . With these definitions we can write (C.25) as

$$\begin{aligned} \delta T_t^{(2)} = \sum_q \sum_{v,w} & \left[(\delta A_v^q)^* K_{vw}^{t(1)} \delta B_w^q + (\delta A_v^q)^* K_{vw}^{t(2)} \delta \bar{B}_w^q \right. \\ & \left. + (\delta B_w^q)^* (K_{vw}^{t(1)})^* \delta A_v^q + (\delta \bar{B}_w^q)^* (K_{vw}^{t(2)})^* \delta A_v^q \right] \end{aligned} \quad (\text{C.29})$$

where

$$K_{v(\sigma_2,\sigma_1,\sigma'_2,\sigma'_1)}^{t(1)} \equiv q_{\sigma_1}^{\sigma'_1} \frac{\partial \left(q_{\sigma_2}^{\sigma'_2} \right)^*}{\partial (A_v)^*}, \quad (\text{C.30})$$

$$K_{v(\sigma_2,\sigma_1,\sigma'_2,\sigma'_1)}^{t(2)} \equiv \frac{\partial q_{\sigma_1}^{\sigma'_1}}{\partial (A_v)^*} \left(q_{\sigma_2}^{\sigma'_2} \right)^*. \quad (\text{C.31})$$

Note that we introduced the two different fluctuations (C.27),(C.28) only because they allow us to write the second-order expansion in a relatively simple form. In fact, these fluctuations are not independent but related through

$$\delta \bar{B}_{\sigma_1,\sigma_2,\sigma'_1,\sigma'_2}^q = (\delta B_{\sigma_2,\sigma_1,\sigma'_2,\sigma'_1}^q)^*. \quad (\text{C.32})$$

Altogether we end up with the following second-order expansion of the Lagrange functional

$$\delta L^{(2)} = \frac{1}{L_s} \sum_q \left(\begin{array}{cccc} \delta \vec{A}^q & \delta \vec{B}^q & \delta \vec{\bar{B}}^q & \delta \vec{\Lambda}^q \end{array} \right)^* \tilde{K}^q \begin{pmatrix} \delta \vec{A}^q \\ \delta \vec{B}^q \\ \delta \vec{\bar{B}}^q \\ \delta \vec{\Lambda}^q \end{pmatrix} \quad (\text{C.33})$$

where

$$\tilde{K}^q \equiv \begin{pmatrix} \tilde{K}^{(A,A)} & \tilde{K}^{(A,B)} & \tilde{K}^{(A,\bar{B})} & \tilde{K}^{(A,\Lambda)} \\ (\tilde{K}^{(A,B)})^\dagger & 0 & 0 & 0 \\ (\tilde{K}^{(A,\bar{B})})^\dagger & 0 & 0 & 0 \\ (\tilde{K}^{(A,\Lambda)})^\dagger & 0 & 0 & 0 \end{pmatrix} \quad (\text{C.34})$$

and

$$\tilde{K}^{(A,A)} \equiv \tilde{K}^{\text{loc}} + \tilde{K}_q^1 + (\tilde{K}_q^1)^\dagger, \quad (\text{C.35})$$

$$\tilde{K}^{(A,B)} \equiv \tilde{K}^{\text{t}(1)}, \quad \tilde{K}^{(A,\bar{B})} \equiv \tilde{K}^{\text{t}(2)}, \quad (\text{C.36})$$

$$\tilde{K}^{(A,\Lambda)} \equiv \tilde{K}^{\text{c}}. \quad (\text{C.37})$$

As described in section 5.4, the antiadiabaticity condition leads to an effective second-order functional only of the density matrix. This condition can be evaluated directly for the second-order expansion (C.33) since the fluctuations $\delta\vec{A}^q$, $\delta\vec{B}^q$, $\delta\vec{\bar{B}}^q$ are some linear functions of the density-matrix fluctuations $\delta\langle\hat{c}_{k,\sigma_1}^\dagger\hat{c}_{k+q,\sigma_2}\rangle$, c.f., Appendix B.2. To this end, we distinguish the fluctuations of the local density matrix $\delta\vec{A}_\rho^q$ and of the variational parameters $\delta\vec{A}_\lambda^q$ as well as the corresponding blocks in the matrix (C.34),

$$\tilde{K}^{(A,A)} = \begin{pmatrix} \tilde{K}_{(\lambda,\lambda)}^{(A,A)} & \tilde{K}_{(\lambda,\rho)}^{(A,A)} \\ (\tilde{K}_{(\lambda,\rho)}^{(A,A)})^\dagger & \tilde{K}_{(\rho,\rho)}^{(A,A)} \end{pmatrix}, \quad \tilde{K}^{(A,B)} = \begin{pmatrix} \tilde{K}_{(\lambda)}^{(A,B)} \\ \tilde{K}_{(\rho)}^{(A,B)} \end{pmatrix}, \dots \quad (\text{C.38})$$

The resulting functional is then given as

$$\delta\bar{L}^{(2)} = \frac{1}{L_s} \sum_q \begin{pmatrix} \delta\vec{A}_\rho^q & \delta\vec{B}^q & \delta\vec{\bar{B}}^q \end{pmatrix}^* \tilde{V}^q \begin{pmatrix} \delta\vec{A}_\rho^q \\ \delta\vec{B}^q \\ \delta\vec{\bar{B}}^q \end{pmatrix} \quad (\text{C.39})$$

with the new kernel

$$\tilde{V}^q \equiv \begin{pmatrix} \tilde{V}^{(A,A)} & \tilde{V}^{(A,B)} & \tilde{V}^{(A,\bar{B})} \\ \tilde{V}^{(B,A)} & \tilde{V}^{(B,B)} & \tilde{V}^{(B,\bar{B})} \\ \tilde{V}^{(\bar{B},A)} & \tilde{V}^{(\bar{B},B)} & \tilde{V}^{(\bar{B},\bar{B})} \end{pmatrix} \quad (\text{C.40})$$

$$= \begin{pmatrix} \tilde{K}_{\rho,\rho}^{(A,A)} & \tilde{K}_\rho^{(A,B)} & \tilde{K}_\rho^{(A,\bar{B})} \\ (\tilde{K}_\rho^{(A,B)})^\dagger & 0 & 0 \\ (\tilde{K}_\rho^{(A,\bar{B})})^\dagger & 0 & 0 \end{pmatrix} - \Delta\tilde{V}^q \quad (\text{C.41})$$

where

$$\begin{aligned} \Delta\tilde{V}^q \equiv & \begin{pmatrix} \tilde{K}_{\rho,\lambda}^{(A,A)} & \tilde{K}_\rho^{(A,\Lambda)} \\ (\tilde{K}_\lambda^{(A,B)})^\dagger & 0 \\ (\tilde{K}_\lambda^{(A,\bar{B})})^\dagger & 0 \end{pmatrix} \times \begin{pmatrix} \tilde{K}_{\lambda,\lambda}^{(A,A)} & \tilde{K}_{\lambda,\Lambda}^{(A,\Lambda)} \\ (\tilde{K}_{\lambda,\Lambda}^{(A,\Lambda)})^\dagger & 0 \end{pmatrix}^{-1} \\ & \times \begin{pmatrix} \tilde{K}_{\lambda,\rho}^{(A,A)} & \tilde{K}_\lambda^{(A,B)} & \tilde{K}_\lambda^{(A,\bar{B})} \\ (\tilde{K}_\rho^{(A,\Lambda)})^\dagger & 0 & 0 \end{pmatrix} \end{aligned} \quad (\text{C.42})$$

Note that \tilde{V}^q (unlike \tilde{K}^q) includes finite couplings also between the fluctuations $\delta\vec{B}^q$, $\delta\vec{\bar{B}}^q$. The calculation of \tilde{V}^q (for fixed \vec{q}) only involves the handling of finite-dimensional matrices. In contrast, the evaluation of the functional (C.25) (instead of (C.29)) would have lead to significantly more complicated equations.

Appendix D. Explicit form of the Gutzwiller-RPA equations

In this appendix, we prove that the general Gutzwiller RPA Eqs. (144) lead to the Green's function matrix (179) if applied to our multi-band Hamiltonian (1). With the abbreviations $\delta D_\mu^q, \hat{D}_\mu^q$ for the three fluctuations $\delta A_v^q, \delta B_w^q, \delta \bar{B}_w^q$ and the corresponding operators $\hat{A}_v^q, \hat{B}_w^q, \hat{\bar{B}}_w^q$, we have to show that the Green's function matrix

$$\Pi_{\mu,\mu'}(\vec{q}, \omega) = \langle \langle \hat{D}_\mu^q; (\hat{D}_{\mu'}^q)^\dagger \rangle \rangle_\omega, \quad (\text{D.1})$$

as given in (179), obeys the equation

$$\delta D_\mu^q = \sum_{\mu'} \langle \langle \hat{D}_\mu^q; (\hat{D}_{\mu'}^q)^\dagger \rangle \rangle_\omega \delta f_{\mu'}^q. \quad (\text{D.2})$$

Using the explicit form (179) of $\tilde{\Pi}(\vec{q}, \omega)$, this equation can also be written as

$$\sum_{\mu'} [1 + \tilde{\Pi}^0(\vec{q}, \omega) \tilde{V}^q]_{\mu,\mu'} \delta D_{\mu'}^q = \sum_{\mu'} \Pi_{\mu,\mu'}^0(\vec{q}, \omega) \delta f_{\mu'}^q \quad (\text{D.3})$$

Note that the excitation amplitudes δf_μ^q enter the problem through the perturbation operator

$$\begin{aligned} \delta \hat{V}_f \equiv \sum_{\mu} \delta f_\mu^q (\hat{D}_\mu^q)^\dagger &\equiv \frac{1}{\sqrt{L_s}} \sum_k \sum_{\sigma_1, \sigma_2, \sigma'_1, \sigma'_2} \hat{c}_{k+q, \sigma'_1}^\dagger \hat{c}_{k, \sigma'_2} \left(\delta f_{\sigma_1, \sigma_2}^{A;q} \delta_{\sigma_1, \sigma'_1} \delta_{\sigma_2, \sigma'_2} \right. \\ &\quad \left. + \delta f_{\sigma_1, \sigma_2, \sigma'_1, \sigma'_2}^{B;q} \epsilon_k^{\sigma_1, \sigma_2} + \delta f_{\sigma_1, \sigma_2, \sigma'_1, \sigma'_2}^{\bar{B};q} \epsilon_{k+q}^{\sigma_1, \sigma_2} \right) \end{aligned} \quad (\text{D.4})$$

which is needed to define the general Green's functions (178).

Before we prove Eq. (D.3), it is instructive to consider the case $\tilde{V}^q = 0$ in which the three fluctuations $\delta A_v^q, \delta B_w^q, \delta \bar{B}_w^q$ are decoupled and we can set $f_w^{B;q} = f_w^{\bar{B};q} = 0$. We start this derivation in the eigenbasis of the Gutzwiller Hamiltonian (180). It leads to the simplest form of the matrix \tilde{E} in Eq. (144) which then reads

$$\begin{aligned} & \left(\omega - (E_{k+q, \alpha_1} - E_{k, \alpha_2}) \right) \delta \langle \hat{h}_{k, \alpha_2}^\dagger \hat{h}_{k+q, \alpha_1} \rangle^{\text{hp/ph}} \\ &= \frac{1}{\sqrt{L_s}} (n_{k, \alpha_2}^0 - n_{k+q, \alpha_1}^0) \delta f_{(k+q, \alpha_1), (k, \alpha_2)}. \end{aligned} \quad (\text{D.5})$$

Here the excitation amplitude is given as

$$\delta f_{(k+q, \alpha_1), (k, \alpha_2)} = \sum_{\sigma_1, \sigma_2} \delta f_{\sigma_1, \sigma_2}^{A;q} (u_{\sigma_1, \alpha_1}^{k+q})^* u_{\sigma_2, \alpha_2}^k. \quad (\text{D.6})$$

Note that the factor $n_{k, \alpha_2}^0 - n_{k+q, \alpha_1}^0 = \pm 1$ in (D.5) represents the particle-hole and the hole-particle channels in Eq. (144). For simplicity, we will drop the corresponding labels hp/ph in the following.

With the transformations (182), (183), Eq. (D.5) leads to

$$\begin{aligned} \delta A_{\sigma_1, \sigma_2}^q &= \frac{1}{\sqrt{L_s}} \sum_k \delta \langle \hat{c}_{k, \sigma_2}^\dagger \hat{c}_{k+q, \sigma_1} \rangle \\ &= \frac{1}{\sqrt{L_s}} \sum_k \sum_{\alpha_1, \alpha_2} (u_{\sigma_2, \alpha_2}^k)^* u_{\sigma_1, \alpha_1}^{k+q} \delta \langle \hat{h}_{k, \alpha_2}^\dagger \hat{h}_{k+q, \alpha_1} \rangle \end{aligned} \quad (\text{D.7})$$

$$= \frac{1}{L_s} \sum_k \sum_{\substack{\alpha_1, \alpha_2 \\ \sigma'_1, \sigma'_2}} \frac{(u_{\sigma_2, \alpha_2}^k)^* u_{\sigma_1, \alpha_1}^{k+q} (u_{\sigma'_1, \alpha_1}^{k+q})^* u_{\sigma'_2, \alpha_2}^k}{\omega - (E_{k+q, \alpha_1} - E_{k, \alpha_2})} (n_{k, \alpha_2}^0 - n_{k+q, \alpha_1}^0) \delta f_{\sigma'_1, \sigma'_2}^q.$$

As expected, we therefore find

$$\delta A_{\sigma_1, \sigma_2}^q = \sum_{\sigma'_1, \sigma'_2} \langle \langle \hat{A}_{\sigma_1, \sigma_2}^q; (\hat{A}_{\sigma'_1, \sigma'_2}^q)^\dagger \rangle \rangle_\omega^0 \delta f_{\sigma'_1, \sigma'_2}^q \quad (\text{D.8})$$

with the ('retarded') Green's function

$$\begin{aligned} & \langle \langle \hat{A}_{\sigma_1, \sigma_2}^q; (\hat{A}_{\sigma'_1, \sigma'_2}^q)^\dagger \rangle \rangle_\omega^0 \\ &= \frac{1}{L_s} \sum_k \sum_{\alpha_1, \alpha_2} \frac{(u_{\sigma_2, \alpha_2}^k)^* u_{\sigma_1, \alpha_1}^{k+q} (u_{\sigma'_1, \alpha_1}^{k+q})^* u_{\sigma'_2, \alpha_2}^k}{\omega - (E_{k+q, \alpha_1} - E_{k, \alpha_2}) + i\delta} (n_{k, \alpha_2}^0 - n_{k+q, \alpha_1}^0) \end{aligned} \quad (\text{D.9})$$

as introduced in (184).

Now we consider the case of a finite interaction matrix \tilde{V}^q . Using our abbreviation δD_μ for the amplitudes δA_v , δB_w , $\delta \bar{B}_w$ the Lagrange functional $\delta \bar{L}^{(2)}$ has the form

$$\delta \bar{L}^{(2)} = \sum_{q, \mu, \mu'} (\delta D_\mu^q)^* V_{\mu, \mu'}^q (\delta D_{\mu'}^q). \quad (\text{D.10})$$

With this additional interaction term, Eq. (D.5) reads

$$\begin{aligned} & (\omega - (E_{k+q, \alpha_1} - E_{k, \alpha_2})) \delta \langle \hat{h}_{k, \alpha_2}^\dagger \hat{h}_{k+q, \alpha_1} \rangle + (n_{k, \alpha_2}^0 - n_{k+q, \alpha_1}^0) \\ & \times \sum_{k', \alpha_3, \alpha_4} U_{k, \alpha_1, \alpha_2}^{k', \alpha_3, \alpha_4}(q) \delta \langle \hat{h}_{k', \alpha_4}^\dagger \hat{h}_{k'+q, \alpha_3} \rangle = \frac{1}{\sqrt{L_s}} (n_{k, \alpha_2}^0 - n_{k+q, \alpha_1}^0) \delta f_{(k+q, \alpha_1), (k, \alpha_2)} \end{aligned} \quad (\text{D.11})$$

where

$$U_{k, \alpha_1, \alpha_2}^{k', \alpha_3, \alpha_4}(q) = \frac{\partial}{\partial \delta \langle \hat{h}_{k+q, \alpha_1}^\dagger \hat{h}_{k, \alpha_2} \rangle} \frac{\partial}{\partial \delta \langle \hat{h}_{k', \alpha_4}^\dagger \hat{h}_{k'+q, \alpha_3} \rangle} \delta \bar{L}^{(2)} \quad (\text{D.12})$$

$$= \sum_{\mu, \mu'} V_{\mu, \mu'}^q \frac{\partial (\delta D_\mu^q)^*}{\partial \delta \langle \hat{h}_{k+q, \alpha_1}^\dagger \hat{h}_{k, \alpha_2} \rangle} \frac{\partial \delta D_{\mu'}^q}{\partial \delta \langle \hat{h}_{k', \alpha_4}^\dagger \hat{h}_{k'+q, \alpha_3} \rangle}. \quad (\text{D.13})$$

and

$$\begin{aligned} \delta f_{(k+q, \alpha_1), (k, \alpha_2)} &= \sum_{\sigma_1, \sigma_2, \sigma'_1, \sigma'_2} (u_{\sigma'_1, \alpha_1}^{k+q})^* u_{\sigma'_2, \alpha_2}^k \left(\delta f_{\sigma_1, \sigma_2}^{A; q} \delta_{\sigma_1, \sigma'_1} \delta_{\sigma_2, \sigma'_2} \right. \\ & \quad \left. + \delta f_{\sigma_1, \sigma_2, \sigma'_1, \sigma'_2}^{B; q} \epsilon_k^{\sigma_1, \sigma_2} + \delta f_{\sigma_1, \sigma_2, \sigma'_1, \sigma'_2}^{\bar{B}; q} \epsilon_{k+q}^{\sigma_1, \sigma_2} \right). \end{aligned} \quad (\text{D.14})$$

The derivatives in (D.13) can be further evaluated using the transformation (182), (183),

$$\frac{\partial (\delta D_\mu^q)^*}{\partial \delta \langle \hat{h}_{k+q, \alpha_1}^\dagger \hat{h}_{k, \alpha_2} \rangle} = \sum_{\sigma_1, \sigma_2} \frac{\partial (\delta D_\mu^q)^*}{\partial \delta \langle \hat{c}_{k+q, \sigma_1}^\dagger \hat{c}_{k, \sigma_2} \rangle} (u_{\sigma_1, \alpha_1}^{k+q})^* u_{\sigma_2, \alpha_2}^k, \quad (\text{D.15})$$

$$\frac{\partial \delta D_{\mu'}^q}{\partial \delta \langle \hat{h}_{k', \alpha_4}^\dagger \hat{h}_{k'+q, \alpha_3} \rangle} = \sum_{\sigma_3, \sigma_4} \frac{\partial \delta D_{\mu'}^q}{\partial \delta \langle \hat{c}_{k', \sigma_4}^\dagger \hat{c}_{k'+q, \sigma_3} \rangle} (u_{\sigma_4, \alpha_4}^{k'})^* u_{\sigma_3, \alpha_3}^{k'+q}. \quad (\text{D.16})$$

Depending on the particular fluctuations δD_μ^q , the remaining derivatives on the r. h. s. of equations (D.15), (D.16) are given as

$$\delta D_\mu^q = \delta A_v^q : \frac{\partial \delta A_{\sigma_2, \sigma_1}^q}{\partial \delta \langle \hat{c}_{k, \sigma}^\dagger \hat{c}_{k+q, \sigma'} \rangle} = \frac{\partial (\delta A_{\sigma_1, \sigma_2}^q)^*}{\partial \delta \langle \hat{c}_{k+q, \sigma}^\dagger \hat{c}_{k, \sigma'} \rangle} = \frac{\delta_{\sigma, \sigma_1} \delta_{\sigma', \sigma_2}}{\sqrt{L_s}}, \quad (\text{D.17})$$

$$\delta D_\mu^q = \delta B_w^q : \frac{\partial \delta B_{\sigma_2, \sigma_1, \sigma'_2, \sigma'_1}^q}{\partial \delta \langle \hat{c}_{k, \sigma}^\dagger \hat{c}_{k+q, \sigma'} \rangle} = \frac{\partial (B_{\sigma_1, \sigma_2, \sigma'_1, \sigma'_2}^q)^*}{\partial \delta \langle \hat{c}_{k+q, \sigma}^\dagger \hat{c}_{k, \sigma'} \rangle} = \frac{\delta_{\sigma, \sigma'_1} \delta_{\sigma', \sigma'_2}}{\sqrt{L_s}} \epsilon_k^{\sigma_1, \sigma_2}, \quad (\text{D.18})$$

$$\delta D_\mu^q = \delta \bar{B}_w^q : \frac{\partial \delta \bar{B}_{\sigma_2, \sigma_1, \sigma'_2, \sigma'_1}^q}{\partial \delta \langle \hat{c}_{k, \sigma}^\dagger \hat{c}_{k+q, \sigma'} \rangle} = \frac{\partial (\bar{B}_{\sigma_1, \sigma_2, \sigma'_1, \sigma'_2}^q)^*}{\partial \delta \langle \hat{c}_{k+q, \sigma}^\dagger \hat{c}_{k, \sigma'} \rangle} = \frac{\delta_{\sigma, \sigma'_1} \delta_{\sigma', \sigma'_2}}{\sqrt{L_s}} \epsilon_{k+q}^{\sigma_1, \sigma_2}. \quad (\text{D.19})$$

With Eqs. (D.12)-(D.19) we are now in the position to evaluate (D.11). To this end, we proceed as in (D.7),

$$\begin{aligned} \delta A_{\sigma_1, \sigma_2}^q &= \frac{1}{\sqrt{L_s}} \sum_k \sum_{\alpha_1, \alpha_2} (u_{\sigma_2, \alpha_2}^k)^* u_{\sigma_1, \alpha_1}^{k+q} \delta \langle \hat{h}_{k, \alpha_2}^\dagger \hat{h}_{k+q, \alpha_1} \rangle \\ &= - \sum_{\mu, \mu'} V_{\mu, \mu'}^q \left\{ \left[\frac{1}{\sqrt{L_s}} \sum_k \sum_{\substack{\alpha_1, \alpha_2 \\ \sigma'_1, \sigma'_2}} \frac{(u_{\sigma_2, \alpha_2}^k)^* u_{\sigma_1, \alpha_1}^{k+q} (u_{\sigma'_1, \alpha_1}^{k+q})^* u_{\sigma'_2, \alpha_2}^k}{\omega - (E_{k+q, \alpha_1} - E_{k, \alpha_2})} (n_{k, \alpha_2}^0 - n_{k+q, \alpha_1}^0) \right. \right. \\ &\quad \times \frac{\partial (\delta D_\mu^q)^*}{\partial \delta \langle \hat{c}_{k+q, \sigma'_1}^\dagger \hat{c}_{k, \sigma'_2} \rangle} \left. \right] \times \sum_{k'} \sum_{\sigma_3, \sigma_4} \frac{\partial \delta D_{\mu'}^q}{\partial \delta \langle \hat{c}_{k', \sigma_4}^\dagger \hat{c}_{k'+q, \sigma_3} \rangle} \delta \langle \hat{c}_{k', \sigma_4}^\dagger \hat{c}_{k'+q, \sigma_3} \rangle \Big\} \\ &\quad + \sum_\mu \langle \langle \hat{A}_{\sigma_1, \sigma_2}^q; (\hat{D}_\mu^q)^\dagger \rangle \rangle_\omega^0 \delta f_\mu^q. \end{aligned} \quad (\text{D.20})$$

The sums over μ and μ' lead to nine contributions which can all be evaluated using Eqs. (D.17)-(D.19). As a result we find

$$\delta A_v^q + \sum_{\mu, \mu'} \langle \langle \hat{A}_v^q; (\hat{D}_\mu^q)^\dagger \rangle \rangle_\omega^0 V_{\mu, \mu'}^q \delta D_{\mu'}^q = \sum_\mu \langle \langle \hat{A}_v^q; (\hat{D}_\mu^q)^\dagger \rangle \rangle_\omega^0 \delta f_\mu^q. \quad (\text{D.21})$$

where the ‘non-interacting’ Green’s functions $\langle \langle \hat{A}_v^q; (\hat{D}_\mu^q)^\dagger \rangle \rangle_\omega^0$ in (D.21) are given as in (D.9), apart from additional factors $\epsilon_k^{\sigma_3, \sigma_4}$ or $\epsilon_{k+q}^{\sigma_3, \sigma_4}$:

$$\begin{aligned} &\left(\begin{aligned} &\langle \langle \hat{A}_{\sigma_1, \sigma_2}^q; (\hat{B}_{\sigma_3, \sigma_4, \sigma'_3, \sigma'_4}^q)^\dagger \rangle \rangle_\omega^0 \\ &\langle \langle \hat{A}_{\sigma_1, \sigma_2}^q; (\hat{\bar{B}}_{\sigma_3, \sigma_4, \sigma'_3, \sigma'_4}^q)^\dagger \rangle \rangle_\omega^0 \end{aligned} \right) \\ &= \frac{1}{L_s} \sum_k \sum_{\alpha_1, \alpha_2} \frac{(u_{\sigma_2, \alpha_2}^k)^* u_{\sigma_1, \alpha_1}^{k+q} (u_{\sigma'_3, \alpha_1}^{k+q})^* u_{\sigma'_4, \alpha_2}^k}{\omega - (E_{k+q, \alpha_1} - E_{k, \alpha_2}) + i\delta} \begin{pmatrix} \epsilon_k^{\sigma_3, \sigma_4} \\ \epsilon_{k+q}^{\sigma_3, \sigma_4} \end{pmatrix} (n_{k, \alpha_2}^0 - n_{k+q, \alpha_1}^0). \end{aligned} \quad (\text{D.22})$$

With (D.21), we have proven the ‘first’ set of Eqs. (D.3), i.e., those with $\mu = v = (\sigma, \sigma')$. If we replace $\delta A_{\sigma_1, \sigma_2}$ in the first line of Eq. (D.20) by

$$\delta B_{\sigma_1, \sigma_2, \sigma'_1, \sigma'_2} = \frac{1}{\sqrt{L_s}} \sum_k \sum_{\alpha_1, \alpha_2} (u_{\sigma'_2, \alpha_2}^k)^* u_{\sigma'_1, \alpha_1}^{k+q} \delta \langle \hat{h}_{k, \alpha_2}^\dagger \hat{h}_{k+q, \alpha_1} \rangle \epsilon_k^{\sigma_2, \sigma_1} \quad (\text{D.23})$$

or by

$$\delta \bar{B}_{\sigma_1, \sigma_2, \sigma'_1, \sigma'_2} = \frac{1}{\sqrt{L_s}} \sum_k \sum_{\alpha_1, \alpha_2} (u_{\sigma'_2, \alpha_2}^k)^* u_{\sigma'_1, \alpha_1}^{k+q} \delta \langle \hat{h}_{k, \alpha_2}^\dagger \hat{h}_{k+q, \alpha_1} \rangle \epsilon_{k+q}^{\sigma_2, \sigma_1} \quad (\text{D.24})$$

the remaining Eqs. (D.3) are derived in the very same way as (D.21). This closes our proof of Eq. (179).

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